



KATHOLIEKE UNIVERSITEIT LEUVEN
FACULTEIT INGENIEURSWETENSCHAPPEN
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AFDELING INSYS
Kasteelpark Arenberg 10 – B-3001 Heverlee – België

SPIN INJECTION IN SEMICONDUCTORS:

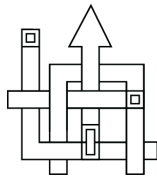
Promotoren:
Prof. Dr. ir. R. Mertens
Prof. Dr. G. Borghs

Proefschrift voorgedragen tot
het behalen van het doctoraat
in de ingenieurswetenschappen

door

Pol VAN DORPE

Maart 2006



in samenwerking met

imec

Interuniversitair Micro-Elektronica Centrum
Kapeldreef 75
B-3001 Leuven (België)



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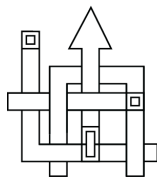
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Spininjectie in halfgeleiders en spintronica in het algemeen werden in de internationale vakpers bij de aanvang van dit doctoraat de hemel in gepraat, spin ging de wereld veroveren. Intussen heeft het de wereld (nog) niet veroverd, maar heeft het mij toch de kans gegeven om boeiende fysische fenomenen grondig te bestuderen. Hierbij speelde het diverse spectrum van menselijke competenties en de fabricatie- en meetmogelijkheden die de labo's van Imec mij verschaften, een bijzonder grote rol in dit onderzoek. Tijdens dit doctoraat heb ik dan ook het genoegen gehad met verschillende mensen te mogen samenwerken die het welslagen van dit doctoraat in hoge mate bevorderd hebben. Zij verdienen allemaal een vermelding in dit dankwoord.

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Pol

Maart 2006

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Abstract

In the field of spintronics the electron spin is used as information carrier in addition to the charge. After earlier successes in metal-based spin-devices, using the spin in semiconductor devices would mean a next integration step. An important challenge for such devices is the creation of a non-equilibrium spin polarization in semiconductors. The most straightforward approach is direct injection of spins from ferromagnetic materials.

In this work we have studied electrical spin injection in GaAs using a variety of tunneling contacts. A measurement of the circular polarization of the light emitted in a light-emitting diode (LED) has been used to determine the injected spin polarization. Using the oblique Hanle effect, a technique that relies on spin precession in an oblique magnetic field, both the magnitude of the injected spin polarization and information about the spin dynamics can be obtained. In order to study spin injection from different tunnel contacts, different spin-LEDs have been designed and fabricated.

Spin-LEDs based on tunnel injection from a sputtered CoFe ferromagnetic metal through an AlO_x -barrier, from an epitaxial MnSb ferromagnetic metal through the Schottky barrier and from a ferromagnetic semiconductor ((Ga,Mn)As) have been shown to result in a high injected electron spin polarization. The polarization persisted up to room temperature for the injector based on the oxide tunnel barrier and a very high polarization has been obtained for the injector based on (Ga,Mn)As at low temperature. Furthermore we have examined and explained the bias dependence of the injected spin polarization for the different devices.

The experimental geometry also allowed to measure the influence of the injected electron spins on the spins of the nuclei in the GaAs lattice. We have investigated the electronic-nuclear spin interactions by examining transient effects upon changing the sign of the injected electron spins and resonant depolarization of the nuclear spins by an alternating magnetic field.

Finally, we have looked into the design of all-electrical spin injection-detection devices. Using self-consistent simulations, we have designed the doping profile for both the injection contacts and for the spin-transport region.

Korte samenvatting

In het spintronica-onderzoeksveld wordt de elektronspin naar voren geschoven als informatiedrager. Na eerdere successen bij metaal-gebaseerde componenten, is het gebruik van de spin in halfgeleidercomponenten een volgende integratiestap. Een belangrijke uitdaging is een elektrische creatie van een spinpolarisatie in halfgeleiders. Een directe injectie van spins vanuit ferromagnetische materialen lijkt hier de meest aangewezen aanpak.

In deze thesis hebben we elektrische spininjectie in GaAs bestudeerd gebruik makende van verschillende tunnelcontacten. Een meting van de circulaire polarisatie van het licht dat uitgezonden werd in een licht-emitterende diode (LED) werd gebruikt om de geïnjecteerde spinpolarisatie te bepalen. Het oblique Hanle effect, een techniek die gebaseerd is op spinprecessie in een schuin magnetisch veld, laat ons toe zowel de spinpolarisatie als informatie over de spindynamica in de halfgeleider te bekomen. Om spininjectie vanuit verschillende tunnelcontacten te meten, werden verschillende spin-LEDs ontworpen en gefabriceerd.

Spin-LEDs gebaseerd op tunnelinjectie van een gesputterd ferromagnetisch metaal (CoFe) door een AlO_x -barrière, vanuit een epitaxiaal ferromagnetisch metaal (MnSb) door de Schottky barrière en vanuit een ferromagnetische halfgeleider ((Ga,Mn)As) resulteerden in een hoge geïnjecteerde spinpolarisatie. Kamertemperatuur spininjectie werd gemeten voor de CoFe- AlO_x -injector en een heel hoge polarisatie werd behaald voor de (Ga,Mn)As-injector bij lage temperatuur. Bovendien hebben we de spanningsafhankelijkheid bestudeerd en verklaard voor de verschillende spin-LEDs.

De experimentele geometrie liet toe de invloed te meten van de elektronspins op de spins van de kernen in het GaAs rooster. We hebben deze spininteracties onderzocht door transiënte effecten te bestuderen na een verandering van het teken van de elektronspins en een resonante depolarisatie van de nucleaire spins in een alternerend magnetisch veld.

Tenslotte hebben we het ontwerp bekeken van volledig-elektrische spininjectie-detectie componenten. Gebruik makende van zelf-consistente simulaties, hebben we het dopingsprofiel ontworpen voor zowel de injectiecontacten als voor het spin-transportgebied.

Curriculum Vitae

November 2, 1978 Born in Leuven, Belgium.

1996-2001 Student at the Katholieke Universiteit Leuven, faculty of applied sciences. Received the M.Sc. degree in electrical engineering, option micro-electronics, in June 2001.

2001-2006 Ph.D research at the Katholieke Universiteit Leuven, faculty of applied sciences, electronics department, and performed at the Interuniversity Micro-Electronics Center (IMEC).

2006 Ph.D. defense: ‘Spin injection in semiconductors’.

Scientific Contributions

Publications

- ‘Highly efficient room temperature spin injection in a metal-insulator-semiconductor light-emitting diode.’ P. Van Dorpe, V.F. Motsnyi, M. Nijboer, E. Goovaerts, V.I. Safarov, J. Das, W. Van Roy, G. Borghs and J. De Boeck. *Jpn. J. Appl. Phys.*, **42**(5B), L502 (2003).
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- ‘Oblique Hanle measurements of InAs/GaAs quantum dot spin-LEDs.’ G. Itskos, E. Harbord, S.K. Clowes, E. Clarke, L.F. Cohen, R. Murray, P. Van Dorpe and W. Van Roy. *Appl. Phys. Lett.*, **88**, 022113 (2006).

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- ‘Spin-dependent phenomena in spin-LED’s, a survey.’ P. Van Dorpe, W. Van Roy, J. De Boeck and G. Borghs. *Proc. SPIE Int. Soc. Opt. Eng.* **5732**, 426 (2005).

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- ‘Design of ferromagnetic tunnel contacts for all electrical spin injection-detection devices.’ P. Van Dorpe, R. Vanheertum, H. Boukari, W. Van Roy and G. Borghs. Presented at: *MMM*, San Jose, USA Nov. 2005.
- ‘Spin injection in semiconductors.’ P. Van Dorpe. Invited Presentation at *the Symposium on nanostructured superconductors*, Leuven, Belgium (2005).
- ‘Spin-dependent transport properties in spin-LEDs, a survey.’ P. Van Dorpe, W. Van Roy, J. De Boeck and G. Borghs. Invited presentation at: *Photonics West (SPIE)*, San Jose, USA, Jan. 2005.
- ‘Spin dynamics in semiconductors’ P. Van Dorpe. Invited lecture at *the Ultrabias summer school*, Biarritz, France, Sept. 2004.

- ‘Strong bias dependence of electron spin injection from (Ga,Mn)As into GaAs.’ P. Van Dorpe, Z. Liu, W. Van Roy, V.F. Motsnyi, G. Borghs and J. De Boeck. Presented at *MRS spring symposium*, San Francisco, USA, Apr. 2004.
- ‘Dynamical nuclear polarization in bulk GaAs due to electrical spin injection from a MnSb tunnel Schottky contact.’ P. Van Dorpe, W. Van Roy, V.F. Motsnyi, G. Borghs and J. De Boeck. Presented at *the MRS spring symposium*, San Francisco, USA, Apr. 2004.
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- ‘Electrical spin injection into a semiconductor.’ G. Borghs, P. Van Dorpe and W. Van Roy. Invited Presentation at *the UK Synchrotron Radiation User Meeting*, Milton Hill, UK, Sept. 2004.
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- ‘Nanomagnets in a semiconductor environment: materials challenges and device perspectives.’ J. De Boeck, P. Van Dorpe, V.F. Motsnyi, W. Van Roy and G. Borghs. Invited presentation at: *International Symposium On Magnetic Materials and Applications*, South-Korea, Dec. 2003.
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- ‘Highly efficient spin injection in ferromagnetic metal - insulator - semiconductor tunnel structures.’ V.I. Safarov, V.F. Motsnyi, J. De Boeck, P. Van Dorpe, W. Van Roy, E. Goovaerts and G. Borghs. Invited presentation at *MRS Fall Meeting Symposium P: Novel Aspects of Spintronic Materials and Devices.*, Boston, USA, Dec. 2002.

Poster presentations

- ‘Nuclear transient effects due to electrical spin injection from a MnSb-Schottky contact in GaAs’ P. Van Dorpe, W. Van Roy, G. Borghs and J. De Boeck. Presented at: *3d Int. Conf. on Physics and Application of Spin Related Phenomena in Semiconductors - PASPS-3.*, Santa Barbara, USA, Jul. 2004.
- ‘Electrical spin injection in GaAs, a survey with different spin injectors.’ P. Van Dorpe, V.F. Motsnyi, Z. Liu, W. Van Roy, G. Borghs and J. De Boeck. Presented at *the Ultrafast Magnetic Switching for Advanced Devices - school*, Leuven, Belgium, Oct. 2003.
- ‘Spin injection from epitaxial NiMnSb into GaAs through a Schottky tunnel barrier.’ W. Van Roy, P. Van Dorpe, V.F. Motsnyi and J. De Boeck. Presented at Spintech II, Bruges, Belgium, Aug. 2003.
- ‘Electrical spin injection in a semiconductor in the MIS heterostructure. Influence of the tunnel barrier, V. Motsnyi, P. Van Dorpe, M. Nijboer, W. Van Roy, J. Das, E. Goovaerts, J. De Boeck, V. Safarov and G. Borghs. Presented at: *26th International Conference on the Physics of Semiconductors - ICPS*, Edinburgh, UK, Jul. 2002.

Acronyms

1D	1-Dimensional
2D	2-Dimensional
2DEG	Two-Dimensional Electron Gas
2DHG	Two-Dimensional Hole Gas
3D	3-Dimensional
AGFM	Alternating Gradient Field Magnetometry
AMR	Anisotropic MagnetoResistance
APD	Avalanche PhotoDiode
BAP	Bir Aronov Pikus
BIA	Bulk Inversion Asymmetry
DMS	Dilute Magnetic Semiconductor
DNP	Dynamical Nuclear Polarization
DP	D'Yakonov-Perel
EHE	Extraordinary Hall Effect
EL	ElectroLuminescence
ESR	Electron Spin Resonance
EY	Elliott-Yafet
FM	FerroMagnet
GMR	Giant MagnetoResistance
HEMT	High Electron Mobility Transistor
IMEC	Interuniversity MicroElectronics Center
IV	Current-voltage
LED	Light Emitting Diode
LHE	Local Hall Effect
LO	Longitudinal Optical phonons
MBE	Molecular Beam Epitaxy
MCD	Magnetic Circular Dichroism

MIS	Metal-Insulator-Semiconductor
MODFET	Modulation Doped Field Effect Transistor
MOSFET	Metal-Oxide-Semiconductor Field Effect Transistor
MR	MagnetoResistance
MRAM	Magnetic Random Access Memory
MTJ	Magnetic Tunnel Junction
NM	Normal Metal
NN	Nearest Neighbour
NMR	Nuclear Magnetic Resonance
OHE	Oblique Hanle Effect
PEM	PhotoElastic Modulator
PL	PhotoLuminescence
RAM	Random Access Memory
RT	Room Temperature
SIA	Structural Inversion Asymmetry
SQUID	Superconducting QUantum Interference Device
STM	Scanning Tunneling Microscope
TEM	Transmission Electron Microscopy
TMR	Tunnel MagnetoResistance

Symbols

Symbol	Description	Units
A	Area	cm^2
B	Magnetic induction	T
B_{loc}	The local field that nuclear spins experience	T
B_N	Effective nuclear field	T
b_N	Nuclear field	T
d	Thickness of the tunnel barrier	cm
$D(E)$	Density of states	cm^{-3}
e	Electron charge ($= 1.6 \cdot 10^{-19}$)	C
E	Electrical field	V/cm
E	Electron energy	eV
E_{ex}	Exchange splitting of the energy bands	eV
E_C	Conduction band	eV
E_V	Valence band	eV
E_g	Energy band gap	eV
E_F	Fermi energy - Fermi level	eV
E_b	Barrier height for a tunnel junction	eV
f	Leakage factor of the nuclear spin polarization	
$F(E)$ or $f(E)$	Fermi-Dirac function, which gives the chance that an energy level E is occupied	
g^*	Landé factor or the effective g-factor	
G	Conductance	S
h	Planck's constant ($= 6.6262 \cdot 10^{-34}$)	J·s
$\hbar$	Planck's constant ($= 2.1091 \cdot 10^{-34}$)	J·s
H_d	Demagnetization field	Am
$\mathbf{H}$	Magnetic Field	A/m

Symbol	Description	Units
I	Current	A
I	The average nuclear spin	Js
I^+, I^-	Intensities of right and left circularly polarized light	W
J	Current density	A/cm ²
J	Total angular momentum	
k	Boltzmann constant ($= 1.38066 \times 10^{-23}$)	J/K
k	Quasi-momentum	1/cm
$k^\uparrow (\downarrow)$	Fermi wave vector in the spin up- (or down) bands	1/cm
L	Orbital angular momentum	
l_{sf}^N	spin diffusion length of a non-magnetic material	cm
M	Magnetization	A/m
m_e^*	Effective electron mass ($= 9.1095 \cdot 10^{-31}$)	kg
m_j	Magnetic quantum number	
M_s	Saturation magnetization	A/m
n	Electron concentration	cm ⁻³
$n_\uparrow (\downarrow)$	Number of spin-up (or spin-down) electrons at the Fermi energy	cm ⁻³
$N_\uparrow$	Density of states for majority electrons	cm ⁻³
$N_\downarrow$	Density of states for minority electrons	cm ⁻³
N_{imp}	Impurity concentration	cm ⁻³
$N_0\alpha$	Exchange splitting of the conduction band	eV
$N_0\beta$	Exchange splitting of the valence band	eV
N_A	Acceptor concentration	cm ⁻³
N_D	Donor concentration	cm ⁻³
P	Degree of circular polarization	%
P_j	Current spin polarization	%
p	Hole concentration	cm ⁻³
r	Vector connecting two electron spins	cm
r_B	Tunnel barrier resistance	$\Omega \times \mu m^2$
r_N	The product of the channel resistivity with the spin diffusion length l_{SF}^N	$\Omega \times \mu m^2$
$R \times A$	Resistance-Area product	$\Omega \times \mu m^2$
S	The spin angular momentum	
S	The average electron spin	Js
S_0	The average injected electron spin	Js

Symbol	Description	Units
t	Time	s
T	Temperature	K or °C
T_1	The longitudinal spin relaxation time	s
T_2	The transverse spin relaxation time	s
T_{1e}	The hyperfine induced nuclear spin relaxation time	s
t_N	The semiconductor channel length	cm
$T(E)$	Tunnel probability as function of the energy	
T_C	Curie temperature	K
T_S	Spin lifetime	s
V	Voltage	V
$v_{\uparrow}$	Fermi velocity for majority electrons	cm/s
$v_{\downarrow}$	Fermi velocity for minority electrons	cm/s
v_0	Volume of a unit cell	cm ³
W	Width of the ferromagnetic electrodes	m
w	Width of the semiconductor channel	m
x	Mn concentration in (Ga,Mn)As	%

Symbol	Description	Units
Δm	Change in magnetic quantum number	
Δ_{so}	Spin-orbit splitting of the valence bands	eV
ΔB	The width of the Hanle curve	T
γ_e	Gyromagnetic ratio of the electron spins	Hz/T
γ_n	Gyromagnetic ratio of the nuclear spins	Hz/T
η	Electron density in a unit cell	cm ⁻³
θ	Angle between the magnetization and the plane	rad
μ_0	Permeability in vacuum ($= 4\pi \cdot 10^{-9}$)	H/cm
μ_I	Nuclear magnetic moment	J/T
μ_B	Bohr magneton	J/T
ν	Frequency of the light	Hz
ξ	Number of nuclei of an isotope in a unit cell	
Π	Spin polarization	%
ρ	Resistivity	$\Omega \cdot \text{cm}$
τ	The electron-hole recombination time	s
τ	Time constant of the nuclear polarization	s
τ_C	The correlation time of a spin-relaxation mechanism	s
τ_{ip}	Momentum relaxation time due to ionized impurity scattering	s
τ_{LO}	Momentum relaxation time due to longitudinal optical phonons	s
τ_P	The momentum relaxation time	s
τ_S	The spin-relaxation time	s
Π	Spin polarization	%
Π_{inj}	Injected spin polarization	%
ϕ	The angle between the magnetic field and the plane	rad
$\Psi(r)$	The electron wavefunction at the nucleus site	
ω	Precession frequency of the electron spins in a random magnetic field	rad/s
Ω	Larmor precession frequency	rad/s
Ω_v	The volume of a unit cell containing 1 Ga and 1 As atom	cm ³

Samenvatting.

Spininjectie in halfgeleiders

S.1 Inleiding

In de klassieke halfgeleider-gebaseerde elektronica wordt de lading van elektronen en gaten aangewend als voornaamste informatiedrager. Transistoren en diodes maken gebruik van elektrische velden om de bandenbuiging in gedopeerde halfgeleiderstructuren aan te passen. Magnetische velden worden zelden gebruikt in halfgeleider-gebaseerde micro-elektronica, in tegenstelling tot data-opslag toepassingen, zoals harde schijven. Bij de laatste toepassing worden magnetische domeinen gebruikt als informatiedragers. Magnetische eigenschappen van materialen zijn gebaseerd op de interactie tussen hun ladingsdragers en hun spin, een intrinsieke magnetische eigenschap van elementaire deeltjes. Deze interactie kan leiden tot nieuwe functionaliteiten, waarbij zowel de spin- als de ladingseigenschappen van elektronische systemen gebruikt worden. In het veld van metaal-gebaseerde magneto-elektronica worden in verschillende toepassingen al spin-afhankelijke effecten op het ladingstransport aangewend. In magnetische multilagen bijvoorbeeld leidt spinafhankelijke verstrooiing tot een groot magnetoweerstandeffect (GMR). Eveneens in magnetische multilagen kan spinafhankelijk tunnelen van ladingsdragers door een isolator die aan weerskanten omgeven is door magnetische lagen leiden tot een groot magnetoweerstandeffect (TMR). Beide effecten hebben geleid tot nieuwe toepassingen, zoals kleinere en gevoeliger magnetische sensoren voor leeskoppen van harde schijven en de ontwikkeling van een magnetisch werkgeheugen (MRAM). Een verdere integratie van spineffecten in de conventionele elektronica impliceert het gebruik van de spin in halfgeleidercomponenten.

Een eerste vereiste voor een dergelijk "spintronische" component, is een efficiënte elektrische generatie van spingepolarizeerde stromen in halfgeleiders, of elektrische spininjectie. De aangewezen methode om spins te injecteren in een halfgeleider lijkt via een ferromagnetisch contact. Een ferro-

magneet heeft een spin-onbalans aan het Fermi-niveau waardoor de stroom door een ferromagneet spingepolariseerd is. Na het aantonen van spininjectie in paramagnetische metalen uit een ohms contact met een ferromagneet, is men dezelfde weg ingeslagen om spininjectie in halfgeleiders te bekomen. Talrijke pogingen om via een ohms contact tussen een ferromagnetisch metaal en een halfgeleider spininjectie te realiseren bleven evenwel zonder resultaat. Uit een theoretische analyse bleek dat de verschillende geleidbaarheid van het metaal en de halfgeleider spininjectie verhinderde. De grootste weerstand in de metaal-halfgeleider combinatie wordt gegeven door de spinonafhankelijke weerstand van de halfgeleider. Een eenvoudige voorstelling van spin-gepolariseerde stromen door twee onafhankelijke spinkanalen, een voorstelling die tot op zekere hoogte gerechtvaardigd is, leert ons dan dat efficiënte spininjectie niet mogelijk is. Verschillende mogelijkheden zijn voorgesteld geweest om dit fundamentele probleem op te lossen, maar de meest succesvolle oplossing is de introductie van een spinafhankelijke weerstand tussen het metaal en de halfgeleider. Een tunnelbarrière lijkt hier een aangewezen oplossing. De tunnelstroom door een halfgeleider is afhankelijk van de toestandsdichtheden aan beide kanten van de tunnelbarrière, die spinafhankelijk is aan de kant van het ferromagnetisch materiaal.

In deze thesis bestuderen we elektrische spininjectie vanuit ferromagnetische materialen, materialen met een natuurlijke spinonbalans aan het Fermi-niveau, in AlGaAs/GaAs-heterostructuren. We fabriceren componenten waarin spininjectie plaatsheeft door tunnel-injectie van spingepolariseerde elektronen vanuit ferromagnetische materialen in halfgeleiders. Om de polarisatie van de geïnjecteerde elektronen te meten, maken we gebruik van de selectieregels voor optische transitie in III-V-halfgeleiders die stellen dat recombinatie van spingepolariseerde elektronen met ongepolariseerde gaten aanleiding geeft tot de emissie van circulair gepolariseerd licht. Door de spin-injector op te nemen in een licht-emitterende diode (LED), zijn we in staat de spinpolarisatie van de geïnjecteerde elektronen te meten. Dit wordt toegepast voor verschillende types van spininjectoren en het laat ons ook toe om de dynamica van de spins in de halfgeleider te bestuderen, net zoals de interactie tussen de geïnjecteerde elektronspins en de spins van de atoomkernen in de halfgeleider. In een laatste deel kijken we verder dan optische detectie en ontwerpen we een component die geschikt is voor elektrische injectie en detectie van elektronspins.

S.2 Spin-LEDs

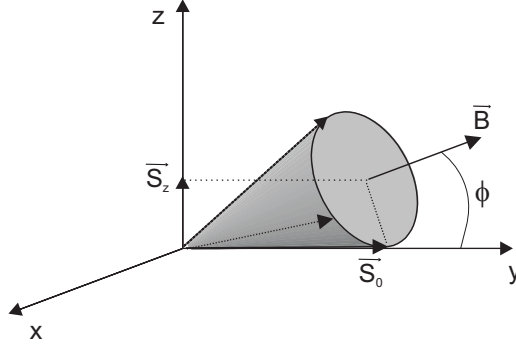
In eerdere onderzoeken naar spininjectie in halfgeleiders is gebleken dat elektrische detectie van een gecreëerde spinpolarisatie lijdt aan parasitaire ef-

fecten die een duidelijke demonstratie van spininjectie onmogelijk maakten. Een alternatieve methode wordt geboden door de selectieregels voor optische transitie in III-V-halfgeleiders. De recombinatie van spin-gepolariseerde elektronen met ongepolariseerde gaten leidt tot de emissie van circulair gepolariseerd licht waarbij de circulaire polarisatie evenredig is met de spinpolarisatie in de richting van de observatie. Een polarisatieanalyse van het geëmitteerde licht kan dus rechtstreeks leiden tot een bepaling van de geïnjecteerde spinpolarisatie. Om een dergelijk effect te kunnen hanteren, moet de spininjector ingebed worden in een lichtemitterende component, zoals een licht-emitterende-diode (een spin-LED). Door een magnetisch contact te fabriceren dat elektronen injecteert in een halfgeleider waar ze met gaten kunnen recombineren, kan men de spinpolarisatie meten van de geïnjecteerde ladingsdragers.

Een moeilijkheid die zich stelt bij de realisatie van een spin-LED, is het feit dat de circulaire polarisatie evenredig is met de spinpolarisatie in de richting van de observatie. De vormanisotropie in ferromagnetische dunne filmen zorgt er echter voor dat de magnetisatie van die filmen in het vlak ligt. Het licht dat gecollecteerd wordt vanuit de bovenkant van een spin-LED plant zich dus loodrecht voort op de magnetisatie van de spininjector en dus ook op de spinpolarisatie in de halfgeleider. In deze configuratie verwachten we dus geen circulaire polarisatie. Men zou dit kunnen oplossen door het licht te meten dat zich parallel met de magnetisatie voortplant, in zijemissie. Deze methode is echter onderhevig aan een aantal randeffecten die een correcte bepaling van de polarisatie bemoeilijken. Deze randeffecten doen zich niet voor in de configuratie waarbij men het licht collecteert dat uit de bovenkant van de spin-LED wordt gecollecteerd. De spins moeten in deze configuratie echter wel gemanipuleerd worden om een loodrechte component te verwerven.

Er zijn twee manieren om de spins te manipuleren: Ten eerste kan de magnetisatie van de ferromagnetische film door een groot loodrecht magnetisch veld uit het vlak getrokken worden. Ten tweede kunnen de spins in de halfgeleider gemanipuleerd worden door een relatief klein magnetisch veld aan te leggen dat niet collineair is met de geïnjecteerde spins. In bepaalde omstandigheden zal de geïnduceerde spinprecessie aanleiding geven tot een loodrechte component van de gemiddelde elektronspin. De eerst vermelde geometrie (de Faraday geometrie) wordt door een aantal onderzoeksgroepen toegepast. Het grote voordeel is de eenvoud van de analyse. Als de magnetisatie gesatureerd is in de richting van het aangelegde magnetische veld, is de circulaire polarisatie de helft van de spinpolarisatie in het geval van een bulk-halfgeleider. Het nadeel van deze techniek is dat er grote velden nodig zijn om de vormanisotropie te overwinnen (rond 2 T). Dit kan additionele randeffecten veroorzaken. De tweede, precessionele techniek heeft het voordeel dat slechts een klein extern magnetisch veld vereist is en dat

de veldafhankelijkheid van de gemeten circulaire polarisatie informatie verschaft over de spindynamica in de halfgeleider.



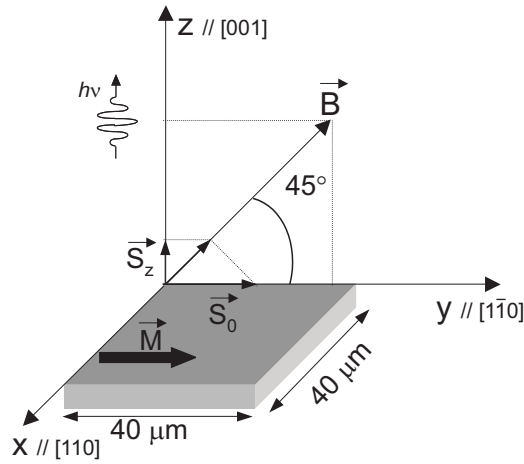
Figuur S.1: Het aanleggen van een schuin magnetisch veld leidt tot spinprecessie, die op haar beurt weer leidt tot een loodrechte spincomponent.

Het is de laatste techniek die we in dit werk toegepast hebben. Door het magnetisch veld in een hoek van 45° met het vlak aan te leggen, veroorzaakt de spinprecessie een loodrechte spincomponent in de halfgeleider. Deze techniek wordt ook het schuine Hanle effect genoemd, of het ‘oblique Hanle effect’ (OHE), naar analogie met het gewone Hanle effect. Bij het Hanle effect wordt de gemiddelde elektronspin gedepolariseerd door het aanleggen van een magnetisch veld dat loodrecht staat op de geïnjecteerde spins. De spinprecessie zorgt ervoor dat enkel de component van de gemiddelde spin die parallel is met het aangelegde veld niet aangetast wordt. Bij het oblique Hanle effect leidt dat tot een loodrechte spincomponent, die gemeten kan worden via de circulaire polarisatie van het licht. Dit principe wordt schematisch afgebeeld in Fig. S.2.

Het resultaat van deze precessie kan ook mathematisch eenvoudig weergegeven worden. In een stationaire toestand vertoont de gemiddelde loodrechte spin, S_Z , de volgende afhankelijkheid van het externe veld:

$$S_Z = S_0 \cdot \frac{T_s}{\tau} \cdot \frac{(\Omega T_s)^2 \cdot 1/2}{1 + (\Omega T_s)^2}, \quad (\text{S.1})$$

met S_0 de gemiddelde geïnjecteerde spin, T_s de spinlevensduur, τ de elektronlevensduur en $\Omega = \frac{g^* \mu_B}{\hbar} \cdot B$, de spinprecessie geïnduceerd door het externe magnetische veld B . g^* , μ_B en $\hbar$ zijn respectievelijk de Landé factor, het Bohr magneton en de constante van Planck. De loodrechte spincomponent stijgt van nul bij het aanleggen van het schuine magnetische veld naar zijn saturatiewaarde bij een hoog magnetisch veld, wanneer de



Figuur S.2: Een schematische voorstelling van de meetgeometrie. Het magnetisch veld staat in een hoek van 45° met het vlak en de magnetisatie. De magnetisatie ligt in het vlak, volgens een van de twee kristalassen $[110]$ of $[1\bar{1}0]$, terwijl het licht uitgezonden wordt volgens de $[001]$ -as. Door de precessie verwerft de gemiddelde spin $\mathbf{S}$ een van nul verschillende loodrechte component $\mathbf{S}_z$.

geïnjecteerde spins verschillende precessie-cycli hebben ondergaan alvorens te recombineren.

Het aanleggen van een schuin magnetisch veld zorgt evenwel voor het optreden van een aantal parasitaire effecten. Door gepaste calibratiemetingen uit te voeren, kunnen die echter volledig geëlimineerd worden.

S.3 Spininjectoren

In dit werk zullen verschillende spininjectoren onderzocht worden. Aluminium oxide tunnelbarrières worden gebruikt om spininjectie in GaAs te realiseren vanuit een gesputterd CoFe topcontact. De natuurlijke Schottky barrière aan het metaal-halfgeleider grensvlak is door middel van de dopering ontworpen om als tunnelbarrière te fungeren tussen een epitaxiaal gegroeid MnSb metaalcontact op GaAs. Tenslotte is spininjectie gerealiseerd in GaAs door een interband-tunnel-proces vanuit de valentieband van de p-type ferromagnetische halfgeleider (Ga,Mn)As naar de conductieband van GaAs.

Deze drie types van spininjectoren zijn geïntegreerd in spin-LEDs. Zowel

de absolute waarde van de geïnjecteerde spinpolarisatie als zijn spanningsafhankelijkheid zullen onderzocht worden.

De verschillende componenten zijn eerst ontworpen door gebruik te maken van zelfconsistente simulaties van de halfgeleiderstructuren in Medici. Deze halfgeleider-simulator laat toe zelfconsistente simulaties uit te voeren van halfgeleider heterostructuren, terwijl het ondermeer zowel tunnel processen door oxidebarrières, Schottky barrières en interband-tunnel-processen in rekening brengt. Deze simulator is gebruikt om zowel vooraf de componenten te ontwerpen, als achteraf om de gemeten spanningsafhankelijkheden te begrijpen.

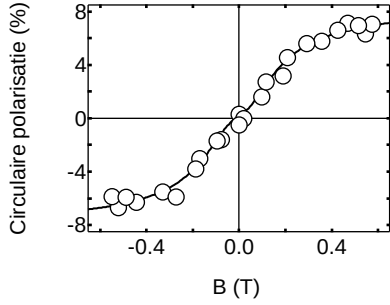
S.4 Spininjectie in metaal-isolator-halfgeleider-combinaties

Deze sectie is gewijd aan de demonstratie van efficiënte spininjectie tot op kamertemperatuur in een metaal-isolator-halfgeleider spin-LED. Voortbouwend op het al bestaande werk in Imec op dit type componenten, zijn we erin geslaagd de geïnjecteerde spinpolarisatie op te krikken van 8% naar 24% op 80 K en deze resultaten uit te breiden naar kamertemperatuur door een optimalisatie van de tunnelbarrière. Voorts hebben we de spanningsafhankelijkheid onderzocht van de geïnjecteerde spinpolarisatie. We hebben twee types spin-LEDs onderzocht, componenten met een ongedopeerd actief gebied en componenten met een p-type dopering in het actieve gebied. De halfgeleider heterostructuur was gegroeid met moleculaire bundel epitaxie. Onmiddellijk na de groei en het breken van het vacuum, werd het sample getransfereerd door de lucht in het sputtersysteem voor de depositie van de tunnelbarrière en de ferromagnetische topelektrode. De oxidelaag werd gedeponneerd in een twee-staps proces. In de eerste stap wordt een dunne Al-laag gesputterd en vervolgens natuurlijk geoxideerd in een gecontroleerde zuurstofatmosfeer bij een druk van 140 Torr. Deze stap wordt vervolgens herhaald. Een dergelijk tweestaps-proces vereenvoudigt een volledige oxidatie van de Al-laag, vermindert de kans op pinholes in het oxide en laat de fabricatie toe van een dikkere barrière. Bij een voorwaartse spanning worden spingepolariseerde elektronen geïnjecteerd vanuit het ferromagnetische contact in de AlGaAs/GaAs heterostructuur. In het 100 nm brede GaAs actieve gebied recombineren ze met gaten die door vanuit p-type substraat aangebracht worden.

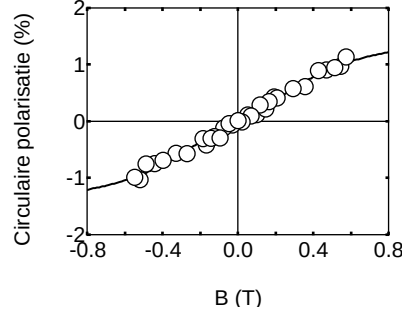
Experimentele resultaten

In figuren S.3 en S.4 wordt de veldafhankelijkheid weergegeven van de circulaire polarisatie van de elektroluminescentie van een spin-LED met een

ongedopeerd actief gebied bij 80 K en bij kamertemperatuur. Een stationaire spinpolarisatie van 24% bij 80 K en van 4.7% bij kamertemperatuur is waargenomen. Dit toont aan dat efficiënte spininjectie plaats vindt, zelfs bij kamertemperatuur.



Figuur S.3: De circulaire polarisatie van de elektroluminescentie in de MIS-spin-LED met een ongedopeerd actief gebied bij 80 K ($\circ$) na correctie voor parasitaire effecten. De volle lijn is een fit volgens de OHE-theorie waaruit een stationaire spinpolarisatie in de halfgeleider van $24 \pm 3\%$ kan afgeleid worden.



Figuur S.4: De circulaire polarisatie van de elektroluminescentie in de MIS-spin-LED met een ongedopeerd actief gebied bij RT ($\circ$) na correctie voor parasitaire effecten. De volle lijn is een fit volgens de OHE-theorie waaruit een stationaire spinpolarisatie in de halfgeleider van $4.7 \pm 1\%$ kan afgeleid worden.

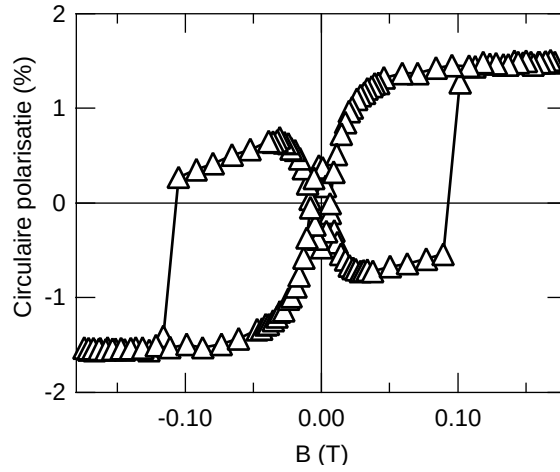
Spanningsafhankelijkheid van de gemeten spinpolarisatie De gemeten spinpolarisatie in de spin-LEDs met een ongedopeerd actief gebied vertoonden een sterke spanningsafhankelijkheid, zowel bij kamertemperatuur als bij 80 K: de gemeten spinpolarisatie daalt sterk bij het verhogen van de spanning. Deze resultaten werden vergeleken met de resultaten van de spin-LEDs met een p-type actief gebied. Hoewel de maximale spinpolarisatie in deze spin-LEDs vergelijkbaar was, viel het op dat in deze LED's geen afhankelijkheid van de geïnjecteerde spinpolarisatie van de aangelegde spanning werd gemeten. Deze spanningsafhankelijkheid van de gemeten spinpolarisatie kan verklaard worden vanuit het raamwerk van het D'yakonov-Perel (DP) spinrelaxatiemechanisme. Door het ontbreken van inversiesymmetrie in GaAs is de conductieband spin-gesplitst. Deze splitsing is sterk afhankelijk van de energie ($\sim E^{\frac{3}{2}}$) en afhankelijk van de $\mathbf{k}$ -vector. Wanneer elektronen met grote energie in de conductieband geëxciteerd (of geïnjecteerd) worden, is die spin-splitsing heel groot. Deze

spin-splitsing werkt als een effectief willekeurig magnetisch veld $\mathbf{B}_{rand}$ waar- rond de elektronenspins een precessie ondergaan. Aangezien dit magnetisch veld willekeurig is, afhankelijk van de $\mathbf{k}$ -vector, zal de gemiddelde spin randomiseren door de verschillende rotaties van de verschillende spins tijdens de thermalisatie. De tijd die de elektronenspins doorbrengen op een $\mathbf{k}$ -vector bij hoge energie is van cruciaal belang. Bij ongedopeerd GaAs wordt die tijd bepaald door energierelaxatie, door de tijd vooraleer een optisch fonon uitgezonden wordt dat het elektron lager in de conductieband doet belanden. Deze tijd is lang genoeg om bij hoge injectie-energie een effectieve randomizing met zich mee te brengen. Bij hoog gedopeerd GaAs wordt de tijd van precessie bepaald door $\mathbf{k}$ -relaxatie door onzuiverheids-verstrooiing. In dit geval is de tijd die het elektron doorbrengt op een $\mathbf{k}$ -positie met hoge energie zo klein dat de effectieve rotatie die de elektronenspins ondergaan uitgemiddeld wordt en de randomizing van de spin tijdens de thermalisatie naar de bodem van de conductieband verhinderd wordt.

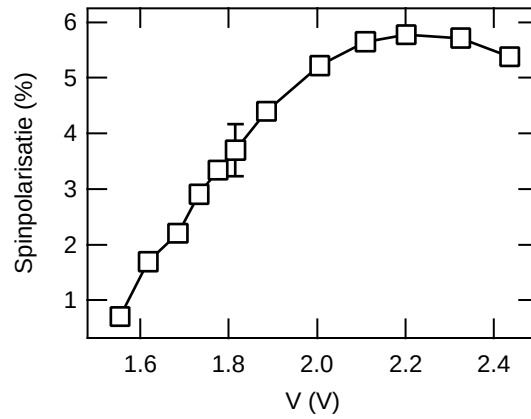
S.5 Spininjectie door de Schottky barrière

De natuurlijke Schottky barrière die gevormd wordt bij het metaal-halfgeleider grensvlak kan door de dopering hoog genoeg op te voeren dun genoeg gemaakt worden opdat tunnelen het dominante transportmechanisme wordt. Aangezien het ferromagnetisch metaal en de halfgeleider direct met elkaar in contact zijn, is de chemische stabiliteit van het grensvlak van primordiaal belang voor een betrouwbare werking van de spininjector.

In dit werk hebben we spininjectie onderzocht vanuit het epitaxiaal gegroeide ferromagnetische metaal MnSb. De halfgeleiderstructuur werd aangepast om direct tunnelen mogelijk te maken tussen het Fermi-niveau van MnSb en de conductieband van AlGaAs. In Fig. S.5 wordt de circulaire polarisatie van het licht vanuit een dergelijke structuur voorgesteld als functie van het externe magneetveld. Een analyse volgens de oblique Hanle effect-theorie leidt tot een stationaire spinpolarisatie van $5,8 \pm 0,2\%$ bij 80 K. De spanningsafhankelijkheid van de gemeten spinpolarisatie wordt weergegeven in Fig. S.8. De gemeten spinpolarisatie is in dit geval 0 bij de drempelspanning voor elektroluminescentie en stijgt gevoelig met de aangelegde spanning. Om dit fenomeen te verklaren, werden zelfconsistente simulaties verricht van het ladingstransport in de structuur. Hieruit bleek dat bij lage spanning de geïnjecteerde elektronen vrij lang verbleven in het spintransport-gebied zodanig dat hun spinpolarisatie al gevoelig verminderd was vooraleer ze het actieve gebied bereikten. Het opvoeren van de spanning zorgde voor een sterke verkleining van de transittijd en een bijhorende stijging van de gemeten spinpolarisatie.



Figuur S.5: De circulaire polarisatie van het licht als functie van het externe magneetveld ($\triangle$) bij 80 K en de fit volgens de oblique Hanle effect-theorie (volle lijn).

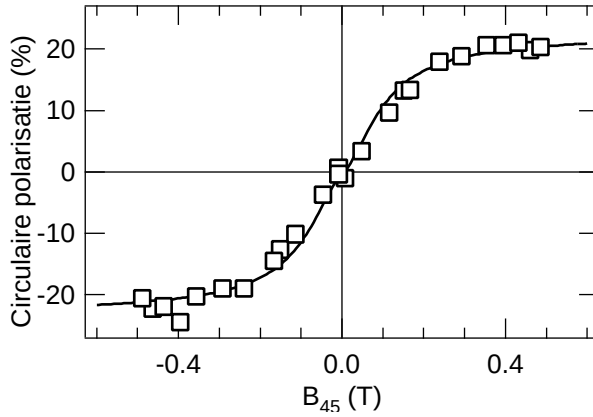


Figuur S.6: De spanningsafhankelijkheid van de gemeten spinpolarisatie in de Schottky spin-LEDs bij 80 K.

S.6 Spininjectie vanuit een magnetische halfgeleider, (Ga,Mn)As

Magnetische halfgeleiders zijn aantrekkelijke kandidaten om een belangrijke rol te spelen in volledig epitaxiaal gegroeide heterostructuren met een spin-

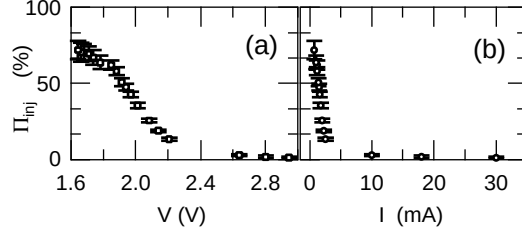
functionaliteit. Het grote nadeel van III-V-gebaseerde magnetische halfgeleiders, zoals (Ga,Mn)As, de best gekende ferromagnetische halfgeleider, is het gatenkarakter van de spin-gepolarizeerde ladingdragers. Gatenspins hebben heel korte levensduren in GaAs door de grote spin-baan-interactie in de valentieband van GaAs. Het is hierdoor bijzonder moeilijk om gatenspins te gebruiken in toepassingen. De gaten kunnen evenwel omgezet worden in elektronen door Zener-tunneling vanuit de valentieband in de conductieband. Dit kan verwezenlijkt worden door een aangepaste dopering te kiezen in een GaAs/(Ga,Mn)As heterostructuur. Door het GaAs hoog n-type te doperen (grootteorde 10^{19} cm^{-3}) is het depletiegebied bijzonder smal zodat bij aanleggen van een achterwaartse spanningsval elektronen gemakkelijk kunnen tunnelen vanuit de valentieband van (Ga,Mn)As in de conductieband van GaAs. Om de spinpolarisatie te meten, hebben we deze structuur verwerkt in een LED-structuur. De (Ga,Mn)As-structuur is 20 nm dik en gegroeid d.m.v. moleculaire bundel epitaxie (MBE). Na de groei en de processtappen om LED-componenten te vervaardigen werd de laag uitgeglod bij een temperatuur van 260 °C gedurende 1 uur. De Curietemperatuur van de magnetische laag steeg vervolgens van 50 K naar 120 K.



Figuur S.7: De circulaire polarisatie van de elektroluminescentie van de (Ga,Mn)As spin-LED bij 4.6 K als functie van het externe magnetische veld ($\square$) en de fit volgens de theorie (volle lijn).

In Fig. S.7 wordt de circulaire polarisatie van de elektroluminescentie weergegeven als functie van het externe magneetveld. De analyse die gebruik maakt van de theorie van het oblique Hanle effect, leidt een geïnjecteerde

spinpolarisatie $\Pi_{inj} = 82 \pm 10\%$, en een stationaire spinpolarisatie in het actief gebied van $= 60 \pm 6\%$. Het blijkt dus dat spininjectoren gebaseerd op magnetische halfgeleiders bijzonder efficiënt kunnen zijn.



Figuur S.8: De geïnjecteerde spinpolarisatie als functie van de spanning (a) en de stroom (b).

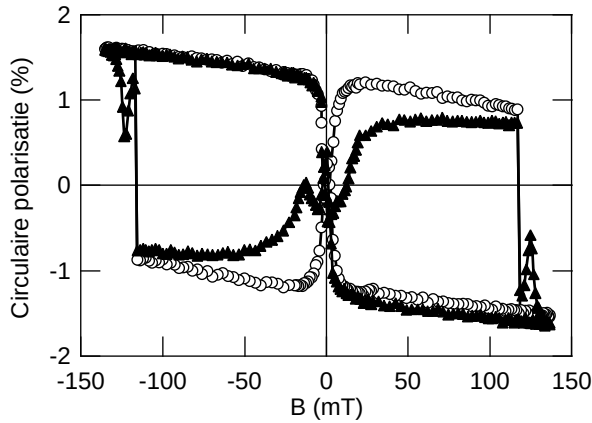
Uit Fig. S.8 blijkt dat de gemeten spinpolarisatie maximaal is bij de drempelspanning voor luminescentie en drastisch daalt bij het verhogen van de spanning. Deze intrigerende spanningsafhankelijkheid hebben we bestudeerd via een duale benadering: In de eerste plaats hebben we zelf-consistente simulaties uitgevoerd van het ladingstransport in de spinLEDs. Deze simulaties boden ons inzicht betreffende de bandenbuiging in de LED, de spatiale distributie van de tunnelstroom en hun spanningsafhankelijkheid. In tweede instantie werden in samenwerking met een onderzoeksgroep in Warschau deze bekomen resultaten gebruikt om de spinpolarisatie van de stroom te berekenen via een ballistisch tunnelmodel (Landauer-Büttiker), uitgaande van een bandenstructuur die verkregen werd door een tight-binding model. Deze duale aanpak stelde ons in staat de spanningsafhankelijkheid van de geïnjecteerde spinpolarisatie te begrijpen. De zelf-consistente simulaties leerden ons al dat het opvoeren van de spanning over de tunneldiode leidt tot het verschuiven van de oorsprong van de tunnelende elektronen van een bulk (Ga,Mn)As-gebied dieper in de depletiezone van de (Ga,Mn)As-GaAs pn-diode. Dit betekent dat bij een hogere aangelegde spanning de elektronen afkomstig zijn vanuit een gebied met een sterk verlaagde gatenconcentratie i.v.m. de bulkconcentratie. Aangezien het ferromagnetisme van (Ga,Mn)As gemedieerd wordt door de interactie tussen de Mn-ionen en de gaten, kunnen de magnetische eigenschappen van de gedepleteerde zone sterk afwijken van de bulk. Bovendien toonden de berekeningen van de spinpolarisatie van de stroom doorheen de tunneldiode eveneens een sterke daling van de polarisatie met de aangelegde spanning. Het bleek dat intrinsieke bandenstructuur-eigenschappen van de pn-diode hiervoor verantwoordelijk waren. In een tunnelproces wordt voldaan aan de

voorwaarde dat de parallelle k -vector van de toestand behouden blijft. Dit zorgt ervoor dat de toestanden van de elektronen uit de minderheidsband zich makkelijker aanpassen aan de toestanden in de conductiebanden. Het opvoeren van de spanning leidt dus tot een effectieve grotere concentratie aan minderheids-spins, wat voor een daling van de spinpolarisatie zorgt.

S.7 Nucleaire spinpolarisatie door de interactie met de geïnjecteerde elektronspins

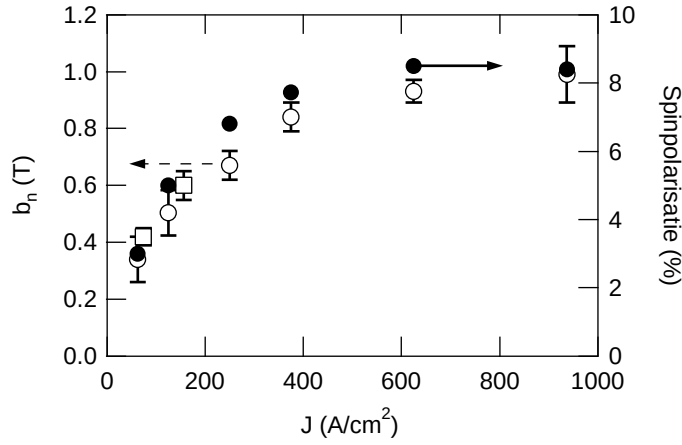
Het is bekend dat een elektronspinpopulatie die uit zijn thermisch evenwicht verkeert, een spinpolarisatie van de kernen van het rooster in de halfgeleider kan veroorzaken via de hyperfijninteractie. Wanneer spins elektrisch geïnjecteerd worden in GaAs, is alvast aan deze voorwaarde voldaan. Om vervolgens een meetbare nucleaire spinpolarisatie te genereren, moeten de processen van nucleaire polarisatie door de interactie met de elektronspins sneller optreden dan de processen van nucleair spinverval. In het geval van hyperfijn-interactieprocessen tussen vrije elektronen en kernspins in GaAs, zijn een vrij grote elektrondichtheid ($\sim 10^{16}$ à 10^{17} cm $^{-3}$) en een lage rooster temperatuur (< 100 K) vereist. Deze dichtheden kunnen optreden bij LEDs met spininjectie doorheen een hooggedopeerde Schottky barrière. In dit type LEDs verloopt het ambipolair ladingstransport meer gebalanceerd dan bij de AlO $_x$ -gebaseerde injectoren, waarbij de gatenstroom domineert. Daarom hebben we de processen van nucleaire polarisatie in detail onderzocht bij de Schottky-injectoren.

De meetmethode die we hanteren, het oblique Hanle effect, is uiterst geschikt voor het bestuderen van de interactieprocessen tussen elektronen en kernspins. De gecreëerde nucleaire spinpolarisatie, die evenredig is met de elektronspinpolarisatie, genereert via de hyperfijninteractie een effectief magnetisch veld dat inwerkt op de elektronspins. Dit effectief veld is altijd parallel met het externe veld en wordt daarbij opgeteld. Daar het oblique Hanle effect gebaseerd is op precessie van elektronspins in een magnetisch veld, zal de verkregen Hanle curve sterk wijzigen indien een effectief veld opgeteld wordt bij het externe veld. Concreet zal het minimum van de Hanle curve, dat normaal optreedt bij 0 T, verschuiven naar de waarde van het externe veld die het nucleaire veld exact compenseert. Voorts treedt er een sterke dip op bij laag veld, wanneer de nucleaire polarisatie verdwijnt als gevolg van de lokale dipool-interacties. In Fig. S.9 wordt de veldafhankelijkheid van de circulaire polarisatie getoond bij 5 K. Uit de stationaire meting ($\circ$) wordt duidelijk dat de Hanle curve bijzonder vernauwd is en evolueert naar een tweede minimum, dat door de beperkte coërciviteit niet kan gevisualiseerd worden. De snelle meting vertoont transiënte effecten bij de doorgang door 0 T en bij het schakelen van de magnetisatie van de

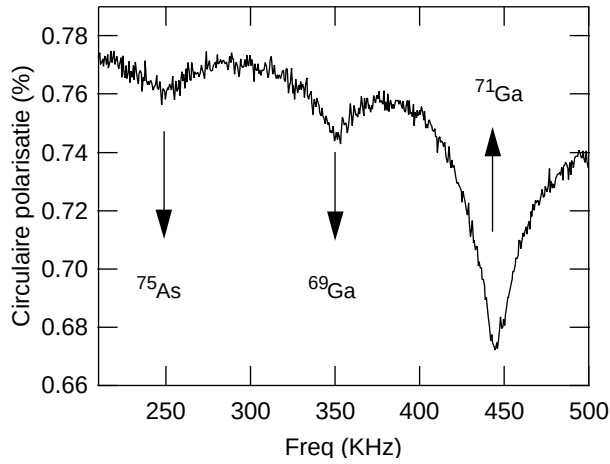


Figuur S.9: De circulaire polarisatie van de elektroluminescentie als functie van het externe veld bij 5 K. (▲): een snelle meting: 1 meetpunt per seconde, (○): een stationaire meting, waarbij een 60 s gewacht werd na het veranderen van het externe veld.

ferromagnetische laag. Deze transiënte effecten zijn het resultaat van veranderingen in de nucleaire polarisatie, veroorzaakt door het schakelen van het veld of van de magnetisatie. Door het intrinsiek trage karakter van de hyperfijninteractie kunnen deze effecten eenvoudig gevisualiseerd worden en aangewend worden om de interacties tussen de kernspins en de elektronspins te bestuderen. Ondermeer de grootte van het effectieve veld dat de kernspins uitoefenen op de elektronspins kan hieruit bepaald worden. In Fig. S.10 staat het nucleaire veld (b_N) weergegeven als functie van de stroomdichtheid (J) in de spin-LED en de spinpolarisatie in het GaAs actieve gebied bij 5 K. Het nucleaire veld stijgt met het verhogen van de stroom door de component en volgt hierbij eenzelfde afhankelijkheid als de elektron-spinpolarisatie, wat in overeenstemming is met de theorie. Door een ac-magnetisch veld aan te leggen dat loodrecht staat op de nucleaire spins en dat resonant is met de Larmor precessiefrequentie van de nucleaire spins in het externe veld, kunnen de nucleaire spins tevens gemanipuleerd worden. Dit wordt duidelijk in Fig. S.11, waar de circulaire polarisatie van de elektroluminescentie staat afgebeeld als functie van de frequentie van het ac-magnetisch veld bij 80 K. De drie dipjes in de polarisatie komen overeen met de resonantiefrequenties van de drie isotopen in GaAs. Bij die frequenties wordt de nucleaire polarisatie van de overeenkomstige isotopen gedeeltelijk vernietigd, wat leidt tot een daling van het effectief veld, en dus een daling van de circulaire polarisatie.



Figuur S.10: Het gemeten effectieve nucleair magnetisch veld (open symbolen) en de elektron-spinpolarisatie in het GaAs actieve gebied (●) als functie van de stroomdichtheid bij 5 K.

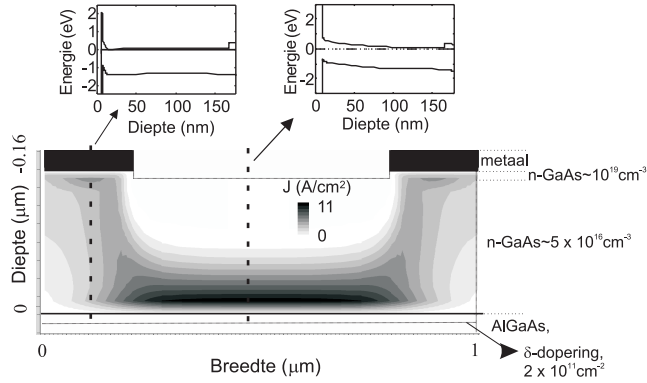


Figuur S.11: De circulaire polarisatie van de elektroluminescentie als functie van de frequentie van het ac-magnetisch veld bij 80 K en 35 mT. De drie GaAs-isotopen zijn duidelijk zichtbaar.

S.8 Elektrische spininjectie en detectie

Hoewel optische detectie van elektrische spininjectie een bijzonder leerzame methode is, die ons toelaat veel informatie te verwerven over het spininjectie-

proces en het gedrag van de spins in de halfgeleider, zal een uiteindelijke toepassing vermoedelijk eerder gebaseerd zijn op elektrische detectie van spins in een halfgeleider. Hoewel er al veel onderzoek op gebeurd is, blijft een duidelijke demonstratie van een elektrische spininjectie en detectie in dezelfde halfgeleidercomponent voorlopig achterwege.



Figuur S.12: De gesimuleerde 2D stroomkarakteristiek van de geoptimaliseerde spin-injectie-detectie component. In de halfgeleider is de stroom geconcentreerd in een regio van ongeveer 40 nm bij het AlGaAs/GaAs grensvlak. De insets tonen de bandenprofielen van doorsneden van de structuur aan het metaalcontact en bij het spintransportgebied.

In deze thesis hebben we het ontwerp van dergelijke spin-injectie-detectie componenten onder de loep genomen, waarbij zowel het ontwerp van de contacten als van het spintransportgebied bekeken werd. Met behulp van 1D en 2D zelfconsistente simulaties hebben we het ladingstransport doorheen een AlGaAs/GaAs-gebaseerde spininjectie-detectie component bestudeerd. Het spintransport gebeurt doorheen de GaAs-laag, terwijl de AlGaAs laag eronder als functie heeft de elektronenstroom te beperken tot een dunne laag (van ~ 100 à 150 nm). We hebben aangetoond dat het doperingsprofiel van de contacten van cruciaal belang is. Ook bij het gebruik van oxide-tunnelbarrières is er immers nood aan een hoge dopering in de halfgeleider dicht tegen het oxide om al bij lage spanning directe tunnelprocessen mogelijk te maken van het metaal naar de halfgeleider. Tevens het doperingsprofiel in de halfgeleider is belangrijk om een optimale distributie van de stroom door het spintransportgebied te bekomen. Met behulp van een delta-dopering in de AlGaAs-laag, dicht tegen de GaAs-actieve laag, kan de ladingsconcentratie in de GaAs laag nauwkeurig bepaald worden. Op Fig. S.12 wordt het gesimuleerde verloop van de stroom getoond doorheen

een AlGaAs-GaAs gebaseerde component. De stroom vloeit doorheen de spintransportlaag, waar de ladingsconcentratie optimaal is. Dit is belangrijk om de spinrelaxatie tijdens het transport te minimaliseren.

In deze thesis hebben we tevens een eerste aanzet gegeven om een spin-injectie-detectie component te fabriceren. Er werd een proces ontworpen om de componenten te fabriceren en eerste karakterisatiemetingen zijn uitgevoerd.

S.9 Besluit

In dit werk hebben we spininjectie bestudeerd in GaAs gebruik makende van verschillende types tunnelcontacten. Met behulp van een optische techniek konden we de spinpolarisatie meten van de geïnjecteerde ladingsdragers door de tunnelinjector op te nemen in een licht-emitterende diode. Een polarisatieanalyse van het licht verschaftte ons informatie over zowel de polarisatie als de dynamica van de geïnjecteerde spins.

Spin-LEDs gebaseerd op tunnelinjectie van een gesputterd CoFe ferromagnetisch metaal door een AlO_X -barrière, vanuit een epitaxiale MnSb ferromagnetisch metaal door de Schottky barrière en vanuit een ferromagnetische halfgeleider ((Ga,Mn)As) resulteerden in een hoge geïnjecteerde spinpolarisatie. Kamertemperatuur spininjectie werd gemeten voor de CoFe- AlO_X -injector en een heel hoge polarisatie werd behaald voor de (Ga,Mn)As-injector bij lage temperatuur. Bovendien hebben we de spanningsafhankelijkheid bestudeerd en verklaard voor de verschillende spin-LEDs.

De experimentele geometrie liet toe de invloed te meten van de elektronspins op de spins van de kernen in het GaAs rooster. We hebben de elektronische-nucleaire spininteracties onderzocht door transiënte effecten te bestuderen en een resonante depolarisatie van de nucleaire spins in een alternerend magnetisch veld.

Tenslotte hebben we het ontwerp bekeken van volledig-elektrische spin-injectie-detectie componenten. Gebruik makende van zelf-consistente simulaties, hebben we het dopingsprofiel ontworpen voor zowel de injectiecontacten als voor het spin-transportgebied.

Chapter 1

Introduction

In classical semiconductor electronics, the charge of electrons and holes is used as primary information carrier. Transistors and diodes exploit the band bending of doped semiconductors upon the application of electric fields to obtain their functionality. Magnetic fields, on the contrary are rarely used in semiconductor electronics, while they are extensively employed in data storage applications such as hard disks, where magnetic domains are used as information carriers. Magnetic properties of materials are based on the interaction of the spin of charge carriers, an intrinsic magnetic property of elementary particles, with their charge. This interaction can lead to novel functionalities, combining both the spin and the charge properties of electronic systems. Indeed in the field of metal based magneto-electronics the spin and the charge properties of electrons are employed to obtain magneto-resistance effects, based on either spin-dependent scattering in magnetic multilayers - the giant magnetoresistance effect (GMR) or on spin-dependent tunneling - the tunnel magnetoresistance (TMR) effect. Both effects have led to new applications such as smaller and more sensitive read heads for hard disks and the development of a magnetic random access memory (MRAM). A further integration of the spin in conventional electronics would implicate the use of spin in semiconductor devices, where the band bending can be accurately controlled by means of doping and electric fields. Furthermore, by band-gap engineering, complicated structures can be fabricated. One step further is bringing the magnetism inside the semiconductor, i.e. creating ferromagnetic semiconductors.

A first prerequisite for an operating spintronic device, is an efficient creation of spin-polarized currents inside semiconductors, or spin injection, by electrical means.

In this work we study electrical spin injection from ferromagnetic materials, materials with a natural spin imbalance at the Fermi level, into

AlGaAs/GaAs heterostructures. We fabricate spin-injection structures using tunnel injection of spin-polarized electrons from ferromagnetic materials into semiconductors. We detect the injected spin polarization in the semiconductor by performing a polarization analysis of the emitted light in a light-emitting-diode. In a second part we look beyond optical detection and design a device suitable for electrical spin injection and detection. We study different (tunnel) injector structures and look into the physics of the spin-transport in these specific structures.

In Chapter 2 we give a brief literature overview dealing with the history of the field of spintronics, and the underlying physics needed for the remainder of this work.

In Chapter 3, we describe in detail the working principle of the spin-light-emitting-diode (spin-LED) and the details of the measurement technique, including the parasitic effects that can occur in the measurement.

In the next three Chapters (Chapter 4, 5 and 6) we fabricate and examine spin-LEDs with three different injector structures, namely a metal-insulator-semiconductor (MIS)-type (oxide tunnel barrier) of injector, a metal-semiconductor (Schottky-tunnel barrier)-injector and a ferromagnetic semiconductor-based (interband-tunneling) injector. In these chapters we describe the experimental results obtained for the different injector structures with regard to the absolute value of the injected spin polarization *and* its dependence on the applied bias voltage. In all three cases we support the interpretation of the bias dependence using self-consistent simulations of the semiconductor structure. Following the experimental results of these chapters, we compare in Chapter 7 the electrical properties of the different types of spin-LEDs and we discuss a source of unpolarized electrons, impact ionization.

In Chapter 8 we study in detail the interaction between electronic and nuclear spins, which results from the injection of spin-polarized electrons in the semiconductor structure. We look at transient effects and nuclear magnetic resonance to understand the phenomena in the devices.

Finally in Chapter 9 we design a device that should be suited for both electrical injection and detection. Using self-consistent simulations we design both the injector structure and the spin-transport region in terms of the doping levels. Finally a processing scheme has been developed and initial characterization experiments have been performed.

Chapter 2

Background

2.1 Ferromagnetism and spin polarization

2.1.1 Ferromagnetic metals

Ferromagnetism arises due to the direct influence of two quantum mechanical effects: spin and the Pauli exclusion principle.

The spin of an electron, combined with its orbital angular momentum, results in a magnetic dipole moment and creates a magnetic field. In a ferromagnetic material the dipoles tend to align spontaneously, without any applied field, which can only be understood from quantum mechanics. According to classical electromagnetism, two nearby magnetic dipoles will tend to align in opposite directions. In a ferromagnet, however, they tend to align in the same direction because of the Pauli principle: two electrons with the same spin state cannot lie at the same position, and thus feel an effective additional repulsion that lowers their electrostatic energy. This difference in energy is called the exchange energy (E_{ex}) and induces nearby electrons to align.

Examples of ferromagnetic materials are iron (Fe), nickel (Ni) and cobalt (Co). They have an only partly filled d-shell, which is split due to the exchange energy, as schematically shown in figure 2.1. In materials with a filled d shell, however, the total dipole moment of all the electrons is zero (e.g. the spins are in up/down pairs).

Spin polarization

The conduction electrons in the 3d ferromagnets are not fully spinpolarized, even when the d-band is strongly spin split, because of the simultaneous

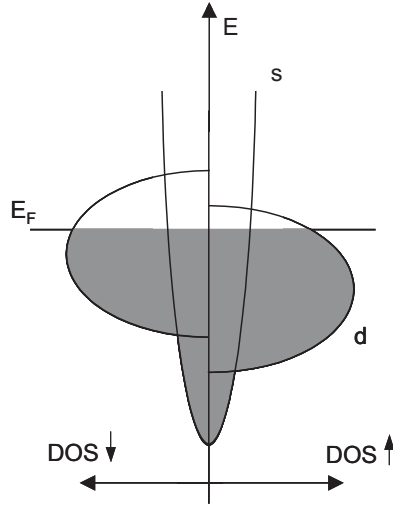


Figure 2.1: The density of states for a typical 3d-ferromagnet. The partially filled d-bands are split by the exchange interaction.

presence of 4s electrons at the Fermi level. The simplest definition of spin polarization is:

$$\Pi = \frac{N_{\uparrow} - N_{\downarrow}}{N_{\uparrow} + N_{\downarrow}} \quad (2.1)$$

with $N_{\uparrow}$ and $N_{\downarrow}$ the density of states for majority and minority electrons at the Fermi level, respectively. A more general, and more useful definition is given by:

$$\Pi_n = \frac{N_{\uparrow} v_{\uparrow}^n - N_{\downarrow} v_{\downarrow}^n}{N_{\uparrow} v_{\uparrow}^n + N_{\downarrow} v_{\downarrow}^n} \quad (2.2)$$

The $\uparrow$ and $\downarrow$ electrons are weighed differently by v , the Fermi velocity. The value of n depends on the type of transport involved. In this case, $n = 0$ for photoemission measurements, $n = 1$ for ballistic transport and $n = 2$ for diffusive transport or tunneling from a ferromagnet to a superconductor through a barrier with low transparency.[1] One has to be careful, though, with applying bulk spin polarization values to heterostructures. The spin polarization of a material in a tunnel structure, e.g., is not a simple property of the ferromagnet alone, but is a joint property of the ferromagnet and the barrier.[2]

Half-metals

Half-metals are defined as ferromagnetic materials that show metallic behavior for spin-up carriers and semiconducting behavior for the spin-down carriers, or in other words, the Fermi level is situated in an energy gap for spin down carriers, while it penetrates a band for spin-up carriers. This leads to a complete spin polarization at the Fermi energy, which is extremely interesting for spintronic applications. NiMnSb, Fe₃O₄, CrO₂ are examples of half-metallic materials. The incorporation of half-metallic materials in spintronic devices has proven to be a challenging task. Especially when the interfacial property is of relevance, such as in a tunneling device, a complete spin polarization at the interface is important. Symmetry breaking, interface states and chemical instability may lead to a deterioration of the interfacial spin polarization, and hence a loss of the half-metallic properties.

2.1.2 Ferromagnetic semiconductors

Making semiconductors ferromagnetic is an intriguing research topic.[3] Controlling both the magnetic and the electrical properties within the same semiconductor really touches the heart of spintronics. Furthermore, controlling the magnetic properties through a bias voltage or an applied current and controlling the resistance through the application of a magnetic field would be useful, if present at room temperature, in a whole range of applications. Indeed semiconductors and insulators can exhibit ferromagnetism. Early studies showed that some Cr-spinels, and rocksalt Eu- and Mn-chalcogenides are ferromagnetic semiconductors, with interesting properties, but however, generally low Curie temperatures and difficult incorporation in standard semiconductor processing.[4] The discovery of ferromagnetism in Mn-doped InAs and GaAs has led to an increased attention for ferromagnetic semiconductors. The Mn-dopant atom (group II from the periodic table) takes the position of a Ga-atom (group III) and hence acts as an acceptor, introducing a hole. Apart from the hole, the Mn ion introduces a magnetic moment as well. The interaction between the Mn-d-states and the valence band p-states (the p-d interaction) leads to a ferromagnetic alignment of the Mn-ions. In this case, as indicated in figure 2.2, the Mn-spin aligns antiferromagnetically with the hole spins surrounding the Mn ion. At high enough Mn- and hole concentrations the Mn doped GaAs layer acts as a ferromagnet. The Curie temperature of this material is directly determined by the Mn concentration (x) and the hole concentration (p) according to the following equation, as derived through the Zener mean field model applied to (Ga,Mn)As: $T_C \sim x \times p^{1/3}$. [5]

Increasing the Mn-concentration, and correspondingly the hole concentration hence leads to a higher Curie temperature. Problematically, the

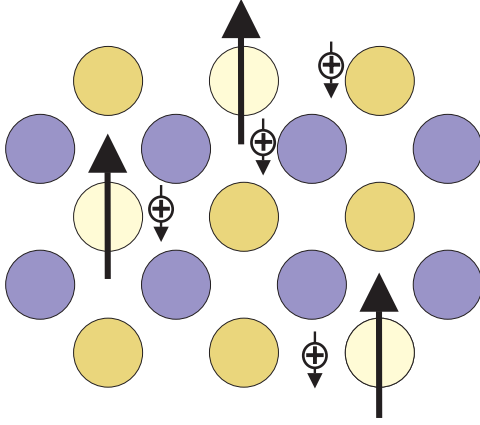


Figure 2.2: Schematic picture of the antiferromagnetic alignment between the Mn- and the hole spins. The hole liquid acts as a glue for the Mn spins.

incorporation of a high number of Mn-atoms in the Ga-sublattice is thermodynamically unstable. Hence, a high-temperature growth of (Ga,Mn)As by Molecular Beam Epitaxy (MBE), which ensures the crystalline quality of the GaAs-phase, leads to the formation of MnAs clusters instead of a homogeneous (Ga,Mn)As film. In order to increase the incorporation of Mn, a non-equilibrium growth scheme is required. This can be provided by low-temperature growth of (Ga,Mn)As. Indeed this leads to the formation of a homogeneous (Ga,Mn)As film, with Curie temperatures ranging from 70 to 110 K [6; 7; 8]. Unfortunately the low-temperature growth leads to the formation of a large concentration of As-antisites, which act as double donors and hence remove part of the holes introduced by the substitutional Mn. Furthermore, there are indications that the incorporation of substitutional Mn is a Fermi-energy limited process[9], which favors the formation of interstitial Mn at high Mn-concentrations. Interstitial Mn also acts as a double donor and is detrimental for the hole concentration and the ferromagnetic properties of (Ga,Mn)As. Recent developments in Nottingham [10] showed that by low-temperature annealing in thin (Ga,Mn)As layers most of the Mn-substitutional atoms diffuse to the sample surface, where they are passivated, by e.g. oxygen [11]. Combined with a precise control of the temperature and the As-flux (to avoid excess As, which gives rise to As anti-sites) this results in strongly reduced compensation, increased hole concentrations and enhanced Curie temperatures. The current T_C record for thin random alloy (Ga,Mn)As-layers is held by the group in Nottingham, as depicted in Fig. 2.3, namely 173 K.[12]

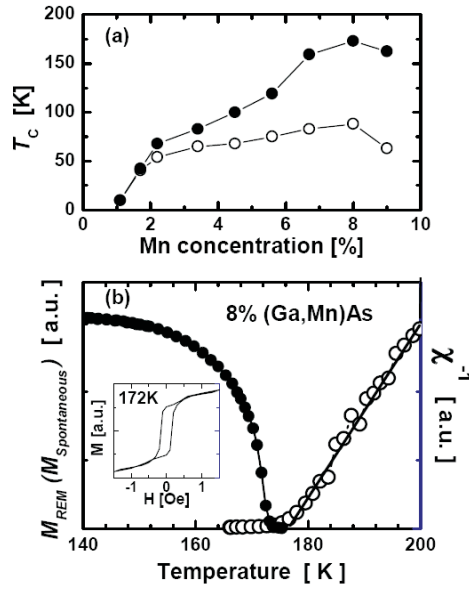


Figure 2.3: (a) The experimentally obtained Curie temperatures in the Nottingham group as function of the Mn concentration before ($\circ$) and after ($\bullet$) annealing. (b) The magnetization data for the record sample, with a Mn concentration of 8%.

Another route towards higher Curie temperatures has been taken by a group of Tokyo University.[13] Instead of growing bulk (Ga,Mn)As, they incorporated a thin delta-doped layer of Mn in a MODFET-like structure (Modulation-doped Field Effect Transistor). Such a structure consists of a heterostructure of a semiconductor and a higher-band-gap semiconductor, such as the AlGaAs/GaAs combination. Introducing the dopant atoms in the higher band-gap material (AlGaAs), creates a two-dimensional electron (or hole) gas in the lower band-gap material (GaAs). Although physically separated from the carriers, the Mn-ions interact with the holes in the two-dimensional hole gas (2DHG). Tanaka optimized the distance of the Mn-delta-doping layer and the 2DHG, resulting in a Curie temperature reaching 250 K.

The hole mediated ferromagnetism in the (III,Mn)V and in some (II,Mn)VI semiconductors provides a direct way of controlling the magnetic properties through changes in the carrier concentration, as has been already demonstrated by means of an electrical bias (for (Cd,Mn)Te[14] and (In,Mn)As[15]) or carrier creation by laser illumination.[16] Experiments on bulk (In,Mn)As

have shown that in the vicinity of the Curie temperature, applying a strong positive gate voltage results in a partial depletion of the (In,Mn)As layer and hence a reduction of the Curie temperature. Also vice versa, a strong negative gate voltage causes accumulation of excess holes and hence an increase of the Curie temperature. Similar results have been obtained in the case of the MODFET-structure of Tanaka and in the case of Mn doped Ge.[17; 18]

2.2 Spin in non-magnetic semiconductors

The total angular momentum ($\mathbf{J}$) of an electron consists of both the electron spin ($\mathbf{S}$) and the orbital momentum ($\mathbf{L}$) : $\mathbf{J} = \mathbf{L} + \mathbf{S}$. Non-magnetic semiconductors provide a nice, clean system for the spin of conduction electrons. Since in zinc-blende or diamond-like semiconductors the conduction band predominantly consists of states with an s-like character, the electrons don't possess any orbital momentum, such that the electron spin is a good quantum number. The valence band, on the contrary consists mainly of states with a p-like symmetry and hence possesses a non-zero orbital momentum ($L = 1$). Furthermore the spin-orbit interaction splits the valence band and pushes the $L = 0$ down in energy. This is depicted in Fig.2.4, where the band structure of GaAs is schematically shown. Most III-V and II-VI semiconductors have a similar band-structure.

2.2.1 Optical selection rules in semiconductors

In bulk III-V or II-VI semiconductors, the selection rules for optical transitions are given by the scheme that is depicted in Fig. 2.5. Illumination of bulk p-type or undoped GaAs with circularly polarized light ($\Delta m = \pm 1$ with an energy $E_g + \Delta_{so} > h\nu \geq E_g$ results in the creation of spin-polarized electron-hole pairs. The holes lose their spin very rapidly (the hole spin-relaxation time is of the order of the momentum relaxation time) and the resulting initial electron spin polarization

$$\Pi = \frac{n^\uparrow - n^\downarrow}{n^\uparrow + n^\downarrow} = 0.5, \quad (2.3)$$

with $n^\uparrow$ and $n^\downarrow$ are the densities of spin-up and spin-down carriers, respectively. Under continuous optical pumping with circularly polarized light, a steady-state spin polarization is created with a polarization $\Pi = 0.5 \frac{\tau_S}{\tau + \tau_S}$, with τ_S and τ the spin relaxation time in the semiconductor and the electron lifetime, respectively. The optically pumped spins are oriented parallel to the exciting light beam.

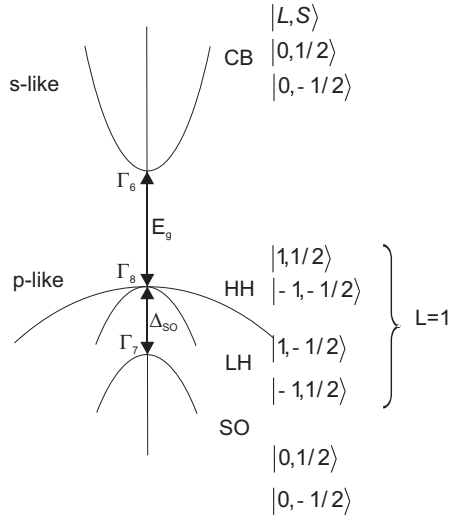


Figure 2.4: The schematic band structure of GaAs, with the conduction band (CB), the heavy hole band (HH) and the light hole band (LH), which are degenerate in the Γ -point and the split-off band (SO), which is Δ_{so} eV separated from the top of the valence band by the spin-orbit interaction. It is clear that the s-like conduction band states have no orbital momentum, while the p-like valence band states have $L = 1$.

For band-edge luminescence, the same selection rules are valid, which implies that the recombination of an ensemble of electron spins with a spin polarization Π in the direction of the quantization axis, results in the emission of circularly polarized light with a polarization of

$$P = \frac{I^+ - I^-}{I^+ + I^-} = 0.5 \times \Pi, \quad (2.4)$$

with I^+ and I^- the intensities of right and left circularly polarized light, respectively.

The selection rules in strained or quantum confined semiconductors. Quantum confinement or strain causes a splitting of the bulk degenerate valence bands. Quantum confinement will push the light hole band more down in energy than the heavy-hole band, while in the case of strain the direction of the splitting depends on the type of strain: compressive or tensile. If the splitting exceeds the thermal energy, the luminescence

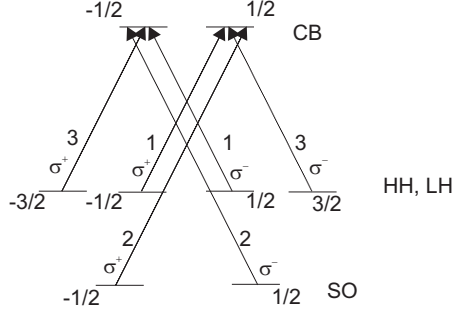


Figure 2.5: The selection rules for interband transitions in III-V and II-VI semiconductors.

will be dominated by heavy-hole or light-hole transitions. This will have a profound influence on the selection rules. Let's consider the case of an unstrained GaAs quantum well surrounded by AlGaAs barriers. In this case the light holes are pushed more down in energy than the heavy holes and the luminescence will be dominated by heavy-hole transitions. Since in the quantum well the heavy holes are pinned along the growth direction, the circular polarization of the luminescence along the growth direction will equal the spin-polarization of the recombining electrons. On the contrary, perpendicular to the growth direction (the case of edge emission), circular polarization is expected to be very small and does not exhibit a straightforward relation to the spin-polarization of the recombining carriers.[19; 20]

2.2.2 Spin dynamics in semiconductors: the Bloch equation

Although spin is an intrinsic quantum-mechanical property of electrons, Bloch realized that we can describe the time-dependence of a spin ensemble using classical motion equations.[19]

We describe the electron spin population by the average electron spin $\mathbf{S} = \sum_{i=1}^n \mathbf{s}_i / n$, with $\mathbf{s}_i$ the spin of the individual electrons and n the amount of electrons. The degree of spin polarization Π in the direction of the average electron spin is $\Pi = 2 \cdot |\mathbf{S}|$.

Upon the application of a magnetic field $\mathbf{B}$ the electron spins, or rather the spin magnetization experiences a torque. This torque will cause the spins to precess at the Larmor frequency

$$\boldsymbol{\Omega} = \frac{g^* \mu_B}{\hbar} \cdot \mathbf{B}, \quad (2.5)$$

with g^* the effective Landé factor and μ_B the Bohr magneton. The precessional motion is altered by damping caused by spin relaxation and the creation or recombination of electrons. Taking these phenomena into account, the evolution of the average spin $\mathbf{S}$ in an external field is described as follows:

$$\frac{d\mathbf{S}}{dt} = \frac{\mathbf{S}_0}{\tau} - \frac{\mathbf{S}}{T_S} + \boldsymbol{\Omega} \times \mathbf{S} \quad (2.6)$$

with $\mathbf{S}_0$ the average injected electron spin, τ the electron lifetime and T_S the spin lifetime. The spin lifetime is defined as ($T_S^{-1} = \tau^{-1} + \tau_S^{-1}$), with τ_S the spin relaxation time.

The Hanle effect.

Using optical pumping with circularly polarized light, a non-equilibrium spin polarization can be created in semiconductors. This induced spin polarization obeys eq. 2.6. Under continuous optical pumping, the application of a transverse magnetic field leads to a depolarization of the spin ensemble due to spin precession in the semiconductor. This effect is called the Hanle effect [19] and is commonly used to determine spin lifetimes in semiconductors. The component of the electron spin parallel to the light beam and perpendicular to the applied field can be described as follows:

$$S_Z = \frac{S_0}{1 + (\Omega T_S)^2} \quad (2.7)$$

2.2.3 Spin relaxation in semiconductors

Spin relaxation is defined as the disappearance of an initial non-equilibrium spin polarization. One can describe spin relaxation as the result of fluctuating random effective magnetic fields. These *effective* magnetic fields typically originate from the spin-orbit or the exchange interaction. The random magnetic field can be described by two parameters: its amplitude, referred to by the average spin precession frequency in the random field, ω and its correlation time τ_C , the time during which the field may be considered as constant. We can consider two limiting cases for the product $\omega \cdot \tau_C$:

a) $\omega \cdot \tau_C \ll 1$: the electron spin undergoes a very small precession during the correlation time τ_C

$$1/\tau_S \sim \omega^2 \cdot \tau_C \quad (2.8)$$

b) $\omega \cdot \tau_C \gg 1$: the electron spin makes many rotations during τ_C

$$\tau_S \sim \tau_C, \quad (2.9)$$

with τ_S the spin relaxation time.

In the case of eq. 2.8 the precession angle during the correlation time is small and the spin undergoes a small angular diffusion. In the case of eq. 2.9, the spin undergoes several precession cycles during the correlation time and hence the spin will be lost in a time of the order of τ_C .

This description is valid for spin relaxation mechanisms where the interaction with the spin is constant during the correlation time, i.e. between scattering events. Another possibility exists when the scattering mechanism itself has a finite probability of reversing the spin and the angle of rotation. In this case the random magnetic field can be seen as a train of delta-functions, corresponding to the scattering events. The correlation time corresponds to the time between the scattering events. Hence, the spin relaxation time is proportional to the correlation time.

Spin relaxation mechanisms

Elliott-Yafet (EY) mechanism. In this mechanism spin relaxation occurs in momentum scattering by charged impurities. The impurity electric field transforms into a magnetic field through the spin-orbit interaction. Hence the spin relaxation rate is proportional to the impurity concentration and inversely proportional to the momentum relaxation time, which corresponds to the correlation time:

$$1/\tau_S \sim N_{imp}, \quad (2.10)$$

This spin relaxation mechanism is not that important in semiconductors, except for high impurity concentrations. It does play a more important role in metals.

D'Yakonov-Perel (DP) mechanism. This spin relaxation mechanism is related to the spin-orbit splitting of the conduction band of semiconductors that lack inversion symmetry, like GaAs (but not Si or Ge). The energy of an electron in the conduction band, should be written in the form:

$$E(\mathbf{k}) = \frac{(\hbar k)^2}{2m^*} + \hbar \mathbf{\Omega}(\mathbf{k}) \cdot \mathbf{S}, \quad (2.11)$$

where $\mathbf{k}$ is the quasi-momentum, $\mathbf{S}$ is the electron spin, and m^* is the effective mass. $\mathbf{\Omega}(\mathbf{k})$ is a vector depending on the orientation of the electron momentum with respect to the crystal axes. The second term is a spin-dependent splitting of the conduction band, which can be seen as an effective

magnetic field. The splitting strongly depends on the electron energy ($\Omega^2 \sim E^3$)

For a given $\mathbf{k}$, $\Omega(\mathbf{k})$ is the spin precession frequency. So the effective magnetic field changes in time because of momentum scattering. Rephrased in terms of the general model of spin relaxation put forward in the beginning of this section, one may state that τ_C is on the order of the momentum relaxation time, τ_P , and $\omega \sim \Omega$. Hence, if τ_P is small:

$$1/\tau_S \sim \Omega^2 \tau_P \quad (2.12)$$

This means that the spin relaxation rate goes down at higher impurity concentrations (when τ_P is smaller). This mechanism is mainly important for high electron concentration and/or high temperature, when the average energy of the electrons increases.

This is the dominating mechanism in most III-V and II-VI semiconductors for low hole concentrations. It is interesting to note that group IV semiconductors, such as Si or Ge, do exhibit inversion symmetry, such that there is no splitting of the conduction band that could lead to the DP-mechanism. Hence the spin lifetime in these systems can exceed the spin lifetimes in III-V semiconductors by orders of magnitude.[21]

Bir-Aronov-Pikus (BAP) mechanism. In p-type semiconductors conduction electrons can lose their spin due to the exchange interaction between the electron and the hole spins.

$$1/\tau_S \sim p \quad (2.13)$$

This mechanism is dominant for conduction electrons in heavily p-doped semiconductors ($p > 10^{18} \text{cm}^{-3}$)

The influence of a longitudinal magnetic field on spin relaxation. The application of a longitudinal magnetic field, i.e. parallel to the direction of the injected spins, typically leads to an increase of the spin relaxation time in bulk semiconductors. The longitudinal magnetic field is superimposed on the random effective magnetic fields that cause spin relaxation. This causes the precession cone to narrow around the external field, which reduces the spin relaxation rate.

Measurements of spin relaxation

Steady-state measurements: the Hanle effect. The Hanle effect has been employed intensively in the seventies and the eighties to determine spin-lifetimes of electrons in the conduction band of III-V-semiconductors.

Monitoring the depolarization of the circular polarization of the photoluminescence in a transverse magnetic field yielded spin relaxation times for different doping levels and different temperatures. This has allowed to investigate which spin relaxation mechanisms dominate in a specific doping/temperature window.

For instance, in figure 2.6 Hanle curves are shown at different temperatures for a p-type doping level of $4 \times 10^{18} \text{cm}^{-3}$. [22] From these Hanle curves the authors deduced that in their samples the BAP-mechanism was dominant in the entire temperature range.

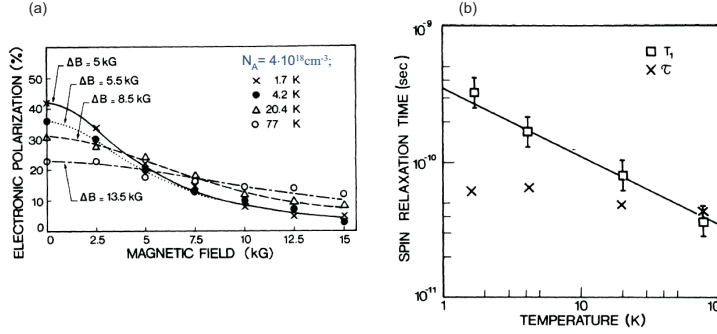


Figure 2.6: (a) Hanle depolarization curves at different temperatures for a p-type doping level of $4 \times 10^{18} \text{cm}^{-3}$, adopted from ref [22]. (b) The temperature dependence of the extracted electron-hole recombination time and the spin-relaxation time.

Time-resolved measurements. Spin relaxation times can also be measured directly, by means of time-resolved methods. The use of ultrafast pulsed lasers allows to create a net concentration of electron spins and to monitor the transient evolution of the spin ensemble. The transient measurement of the spin-status can be performed by various means. Quite a lot of interesting experiments have been performed using pump-probe schemes employing the use of the Faraday or the Kerr rotation to measure the magnetization state of the electron ensemble.[23] This scheme is particularly useful in n-type samples, where the photogenerated holes rapidly recombine with the electrons, resulting in a partial spin polarization of the majority carriers. Upon the application of a magnetic field, with a component perpendicular to the average injected spin, the injected spins precess around the field. The Faraday or the Kerr effect allows to monitor this by measuring the rotation of the linear polarization of the transmitted (Faraday) or

the reflected (Kerr) probe beam. Using this technique, spin lifetimes up to 100 ns have been measured.

Another possible route is to measure the transient behavior of the circular polarization of the photoluminescence after a short pulse of a circularly polarized laser beam. This scheme is more appropriate for systems where the photogenerated electrons are minority carriers.

Example: low temperature spin relaxation in n-type GaAs. Both time-resolved and Hanle measurements have been employed to study spin relaxation in n-type GaAs.[24; 23] The results of these experiments are summarized in figure 2.7. It is striking that the highest spin relaxation time is reached for electrons in the donor band for doping levels near the metal-insulator-transition, i.e. with doping levels between $1 \times 10^{16} \text{cm}^{-3}$ and $1 \times 10^{17} \text{cm}^{-3}$. As can be seen on the graph, spin-lifetimes up to 200 ns have been measured. At higher donor concentrations, the spin relaxation time steeply decreases, following a power law as function of the donor concentration. At smaller doping levels there is also a decrease, but less dramatic.

The mechanisms can be explained as follows: at small donor concentrations and at low temperature, the electrons are localized at the donor sites and experience a strong coupling with the nuclear spins in the environment of the donor atom. This causes the spin relaxation to be relatively efficient. With increasing impurity concentration the electron wave functions start to overlap and the exchange interaction between neighboring donor electrons causes flip-flop transitions of the electron spins. This results in a rapid transfer of the spin over the different donor sites such that the effect of the individual nuclear spin fluctuations decreases. This causes τ_C , the correlation time of this spin-relaxation mechanism, to decrease and the spin relaxation experiences motional narrowing. Above the metal-insulator transition the DP-mechanism will start to dominate the spin-relaxation. An increase of the donor concentration will cause an increase of the Fermi energy, and hence of the wave vector k , such that the spin relaxation time decreases.

2.2.4 Nuclear spin polarization

The nuclear spin system in semiconductors.

Apart from the electronic spin systems, most semiconductors possess their own nuclear spin system. GaAs, for instance, has three nuclear isotopes: ^{69}Ga , ^{71}Ga and ^{75}As with a natural abundance of 60%, 40% and 100%, respectively. All of these isotopes have a nuclear spin $I = 3/2$. [25; 26] Upon the application of a magnetic field $\mathbf{B}$, the levels of the individual spins split, into different states with values along the direction of the magnetic field

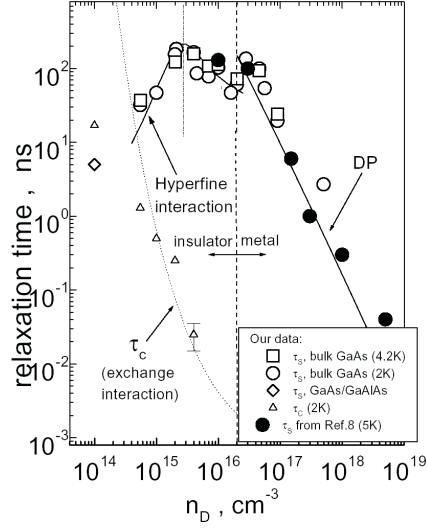


Figure 2.7: The spin-relaxation times as function of the Si donor concentration in GaAs at low temperature.

of $m = -3/2, -1/2, 1/2$ and $3/2$. The Zeeman energy of these individual states equals $E = -m\hbar\gamma B$, with γ the gyromagnetic ratio of the individual isotopes. This energy is very small compared to the thermal energy for moderate values of B and generally leads to a very low thermal polarization.

The hyperfine coupling between electron and nuclear spins.

The creation of a non-equilibrium spin polarization in semiconductors can lead to a significant change of the spin polarization of the nuclear spin system in the semiconductor.

The hyperfine interaction between electron and nuclear spins is known to transfer polarization from one spin system to the other in the presence of a longitudinal magnetic field. Especially in the case when one spin system is strongly out of equilibrium, the other spin system can be polarized very efficiently. In optical pumping experiments, a steady-state spin polarization is generated in the electron system. Compared to the thermal spin-polarization, which is negligible at the moderate fields typically used in these experiments, this represents a strong non-equilibrium situation. Under the proper conditions, this can lead to a strong polarization of the nuclear spin system.

There are two types of hyperfine interaction: the dipolar hyperfine inter-

action and the contact hyperfine interaction. The dipolar hyperfine interaction is an interaction between the magnetic moments of the electron and nuclear spin in analogy to the classical dipolar interaction between magnetic moments. The energy of this interaction is written as:

$$\mathcal{H}_{dip} = \frac{1}{r^3} \gamma_n \gamma_e \hbar^2 \left(\mathbf{S} \cdot \mathbf{I} - 3 \frac{(\mathbf{S} \cdot \mathbf{r})(\mathbf{I} \cdot \mathbf{r})}{r^2} \right) \quad (2.14)$$

with $\mathbf{r}$ the vector connecting the two spins, γ_n and γ_e the gyromagnetic ratios of the nuclear and the electron spins, respectively.

The contact hyperfine interaction becomes important when there is a finite density of the electron spin on the nucleus site. The energy of its interaction is written as:

$$\mathcal{H}_{cont} = -\frac{8\pi}{3} \gamma_n \gamma_e \hbar^2 |\psi(\mathbf{r})|^2 \mathbf{S} \cdot \mathbf{I} \quad (2.15)$$

with $|\psi(\mathbf{r})|$ the electron wavefunction at the nucleus site.

In most semiconductors, such as GaAs, the conduction states near the Γ -point have an s-like character and have their highest probability inside the nucleus. Hence, for conduction electrons, the contact hyperfine interaction is the dominant type. In the valence band the states have a p-like character, which causes the contact hyperfine interaction to be negligible. In this case the dipolar hyperfine interaction dominates.

It has been shown already in 1968 by Lampel that optical pumping of Si could lead to an enhanced nuclear spin polarization.[27] A few years later Ekimov and Safarov showed that the polarization of the nuclear spin system could not only be greatly enhanced by optical pumping in an AlGaAs-alloy, but also that the induced nuclear spin polarization acts back on the electron spins as an effective magnetic field.[28] This effective field is also known as the Overhauser shift (the shift of the electron spin resonance (ESR) field by the presence of a nuclear spin polarization).

Due to the long transverse spin relaxation time of the nuclear spins, in steady-state conditions the nuclear field is parallel to the external field. The nuclear field is superimposed on the external field and can manifest itself in several ways. Under optical pumping in a longitudinal magnetic field, the induced nuclear field can be either parallel or anti-parallel to the external field, depending on the helicity of the incident light. A parallel alignment can cause an additional suppression of the spin-relaxation due to the enhanced total field, while an anti-parallel alignment reduced the total field and hence can accelerate spin relaxation. This is the technique used by Berkovits *et al* to demonstrate nuclear spin polarization in GaAs.[29] A direct measure of the nuclear field can be provided by the Hanle effect, where the magnetic field depolarizes the electron spins. In this geometry, however, the applied field is perpendicular to the external field, which makes

it impossible to create a nuclear polarization. D'yakonov and coworkers, however, showed in 1974 that the application of an oblique field makes it possible to visualize the nuclear field in a Hanle-type experiment.[30] They called it the oblique Hanle effect. The transverse component of the field causes the depolarization, while the longitudinal causes the creation of a non-equilibrium nuclear spin-polarization. Since the nuclear field is parallel to the external field, the nuclear polarization causes a shift of the Hanle curve, the size of the shift being the nuclear field.

Nuclear spin polarization in time-resolved measurements.

In the previous paragraph, the nuclear spin polarization was created and measured using steady-state techniques, i.e., using continuous wave (cw)-laser illumination of the sample and a steady-state measurement of the circular polarization of the light. More recent experiments in the group of David Awschalom in Santa Barbara (UCSB) have been performed using time-resolved techniques for the creation and the measurement of spin-polarized charge packets in semiconductors. Using ultrafast pumped lasers, they are able to monitor the transient behaviour of the optically injected spins. In the presence of nuclear spin polarization, the Larmor precession frequency changes due to the superposition of the nuclear field and the external field.[31] Furthermore, the periodic generation of spin and charge packets due to the laser pulse train generates a hyperfine tipping field that brings the nuclear spins into resonance.[31; 32; 33] By examining the fine-structure of the resonance peaks, they were able to distinguish both $\Delta m = 1$ ($\frac{-3}{2} - \frac{-1}{2}$, $\frac{-1}{2} - \frac{1}{2}$ and $\frac{1}{2} - \frac{3}{2}$) transitions and $\Delta m = 2$ ($\frac{-3}{2} - \frac{1}{2}$ and $\frac{-1}{2} - \frac{3}{2}$) transitions. Although $\Delta m = 2$ -transitions are generally forbidden for GaAs, they have been observed in this experiment. These transitions are linked to the quadrupolar interactions of the nuclear spins with local electric fields. In AlGaAs, e.g., the random replacement of Ga atoms by Al atoms generates local electric fields, generating a strong quadrupolar interaction. In the pump-probe experiments, the periodic excitation of charge packets generates local electric fields that seem to be responsible for the observed effects.

2.3 Spin injection

Electrical spin injection in non-magnetic materials is a research topic with a rich history. Already in the early seventies, Tedrow and Meservey examined the spin-polarization of ferromagnetic materials by tunnel spectroscopy of ferromagnet/oxide/superconductor tunnel junctions in a large in-plane magnetic field.[34] Later Aronov proposed that electrical spin injection should be possible into a wide range of non-magnetic materials, such as metals, su-

perconductors and semiconductors.[35; 36; 37] Spin injection in metals has been successfully experimentally demonstrated in the eighties by Johnson and Silsbee,[38] even before the discovery of the giant magnetoresistance effect (GMR) in ferromagnet/normal metal multilayers by Fert et al.[39] This discovery, together with the demonstration of the tunnel-magnetoresistance (TMR) effect at room temperature by Moodera et al.[40] triggered the magnetic community to investigate in detail spin-polarized currents in non-magnetic systems, such as metals *and* semiconductors.

2.3.1 Spin injection in metals

The seminal Johnson-Silsbee experiments.

The exploration of spin injection in metals has been initiated by Johnson and Silsbee.[38] They showed clear evidence of the creation of a spin-polarized current in bulk aluminum by injection from a $\text{Ni}_{0.8}\text{Fe}_{0.2}$ contact. They employed a non-local technique to probe the spin-split density of states in the aluminum using a second ferromagnetic voltage probe. Utilizing the Hanle effect, they demonstrated the diffusion and the precession of spins from the injection to the detection contact. In the non-local scheme the current path and the ferromagnetic voltage probe are separated. Hence, the ferromagnetic contact probes the splitting of the spin dependent quasi-Fermi levels due to spin diffusion from the injection contact. A schematic picture is shown in Fig. 2.8. The injected electron spins cause a splitting of the spin-dependent quasi-Fermi levels in the non-magnetic metal. A second ferromagnetic contact, that is connected to the ground through a voltage meter with infinite impedance will acquire a voltage drop V_S with respect to the normal metal, that depends on the induced magnetization in the normal metal and the spin-dependence of the normal metal-ferromagnetic metal contact.

In order to have an unambiguous proof of the origin of their spin-signals, they applied a magnetic field $H_\perp$ that is perpendicular to the magnetization of the injector and the detector electrodes and causes the spins to precess in the metal. At high enough fields, this causes a dephasing of the injected spin polarization and the spin-signal vanishes. This is shown in Fig. 2.9, where the spin signal clearly vanishes with the applied field.

Giant magnetoresistance effect (GMR).

The giant magnetoresistance (GMR) effect was discovered in the late eighties by two independent research groups: Peter Gruenberg's group of the KFA research institute in Jülich, Germany[41], and Albert Fert's group of the University of Paris-Sud[39]. They observed very large resistance changes

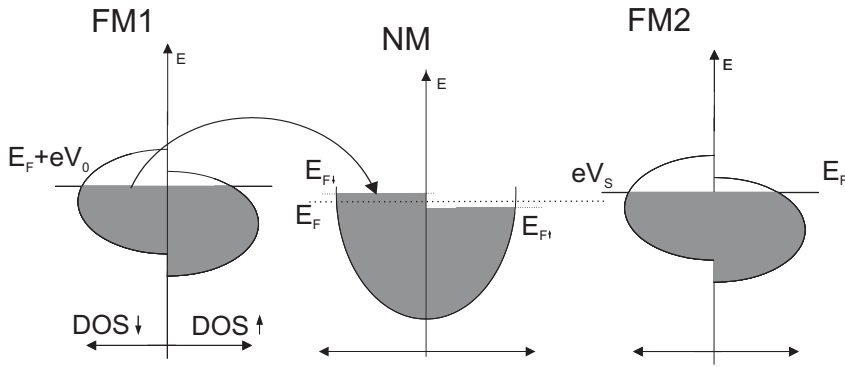


Figure 2.8: A simple picture of the non-local detection of spin-accumulation in the Johnson-experiment.

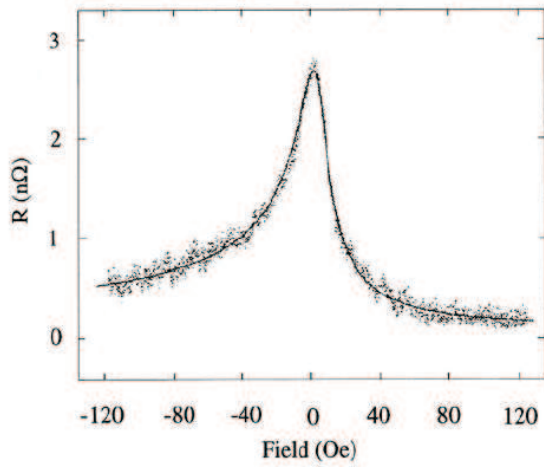


Figure 2.9: A measured Hanle depolarization curve, together with the fit of the non-local spin signal in the spin-injection structure of Johnson.[38]

– 6 percent and 50 percent, respectively – in materials comprised of alternating thin films of magnetic and non-magnetic metals. The basic mechanism behind these large magnetoresistance effects is spin-dependent scattering in magnetic layers with a non-collinear magnetization, the non-magnetic metal serving as a spin-transport region and allowing a decoupling of the magnetizations of the individual magnetic layers. Their results created large

attention and quickly magnetic multilayers with only few layers were optimized for optimum magnetoresistance and sensor characteristics. Reducing the amount of layers to a basic ferromagnet/normal metal/ferromagnet-structure leads to a device that is generally known as a spin-valve. Optimized devices (materials combinations and layer thickness) lead to magnetoresistance effects up to 20% at room temperature in low external fields, making these devices ideal for sensor applications.

The tunnel magnetoresistance effect (TMR).

Already in 1975 Jullière discovered that a tunnel junction comprised of two magnetic metals separated by a thin insulating tunnel barrier exhibit a magnetoresistance effect.[42] In his experiment an Fe layer and a Co layer were separated by a Ge-tunnel barrier. At low temperature (4.2 K) a magnetoresistance effect of 14% was observed. Jullière proposed a simple model to explain his results based on the bulk spin polarization of the involved ferromagnetic layers. According to his model, the tunnel magnetoresistance effect can be described by the following equation:

$$\Delta R/R = \frac{R_a - R_p}{R_a} = \frac{2P_1P_2}{1 + P_1P_2}, \quad (2.16)$$

with R_a and R_p the resistance of the tunnel junction in the case the magnetization of the magnetic layers are anti-parallel or parallel aligned, respectively. P_1 and P_2 correspond to the bulk spin polarizations at the Fermi energy in the two magnetic materials. This model is based on simple density-of-states considerations and the magnetoresistance depends therefore only on the parameters of the ferromagnetic layers.

Following the discovery of the GMR-effect, the interest in TMR revitalized and in 1995 Moodera could demonstrate for the first time a large magnetoresistance effect at room temperature. In CoFe/AlO_x/Co-tunnel junctions a TMR-effect of 11.8% was measured at room temperature.[40]

The prospect of using magnetic tunnel junctions (MTJ) exhibiting the TMR-effect as non-volatile memory elements in a Magnetoresistive Random Access Memory (MRAM), generated a large worldwide research effort to boost the TMR and to understand its properties in more detail.

Especially the preparation of the tunnel barrier has become an intensive area of research. Initially, aluminum oxide gave the best results, and a wide range of oxidation techniques were employed, such as natural oxidation, plasma oxidation, UV-assisted oxidation and oxidation in an ozon environment.[43] The largest TMR-effects based on AlO_x-barriers have been reported with plasma-oxidized barriers, currently up to 70%.[44] A drawback of the method using plasma-oxidation is the very high resistance-area product (RA) of these tunnel junctions (up to MΩ · μm²), while in

MRAM typically a low RA is required. Natural oxidation has been shown to result in a lower RA, but exhibits a lower TMR and more extrinsic defects.

The type of barrier also plays an important role regarding the sign and the magnitude of the spin polarization of the electrodes. It appeared that, although the Jullière equation seemed to provide a good prediction of the TMR, several issues remained unresolved. The sign of the bulk spin polarization of the ferromagnetic materials didn't correspond with the experimentally obtained sign for tunnel junctions. Furthermore, Teresa *et al* showed that using a different type of barrier may result in a different sign of the magnetoresistance, indicating a change in the sign of the spin polarization.[2] It is now well established that the type of bonds that are formed at the ferromagnet/oxide interface can change the sign of the interfacial spin polarization.[45; 46] Since tunneling is a particularly interfacial property, the spin polarization to be used in the Jullière equation is the spin polarization at the ferromagnet/oxide interface.

Aluminum oxide is an amorphous tunnel barrier and it is generally assumed that tunneling through an amorphous tunnel barrier does not conserve the in-plane wavevector, masking possible band-structure effects in the tunnel barrier. It has been predicted, however, that at the Fermi energies of bcc Fe, bcc Co and CoFeB, there is a difference in the symmetries of wave functions between the majority and minority spin channels. Specifically there is a Δ_1 Bloch state for the majority, but not for the minority spin channel. For some crystalline insulating materials, states with this Δ_1 symmetry will decay much more slowly than states with different symmetries. More specifically, a very large magnetoresistance has been predicted in a Fe/MgO/Fe-epitaxial magnetic tunnel junction based on this effect.[47; 48] Indeed, recently several groups have demonstrated very large magnetoresistance effects (200% up to 355% at room temperature) using magnetic tunnel junctions based on MgO with a (001)-orientation.[49; 50; 51] Furthermore, this effect persists at room temperature and yields magnetic tunnel junctions with a very low RA, making them very attractive for MRAM-applications.

Spin injection in mesoscopic ferromagnetic-normal metal structures

Recently the early results of Johnson and Silsbee on bulk single crystal metals have been extended to thin films and nano-dimensions by Jedema *et al*. [52; 53] They also utilized a non-local measurement technique to show unambiguously spin injection in Cu and Al, furthermore they increased their signal by using an AlO_x -tunnel barrier between the ferromagnetic metal and the non-magnetic metal, Al in this case.

2.3.2 Spin injection in semiconductors: the conductance mismatch

Based upon the success of GMR in all-metallic structures and the success of charge based semiconductor electronics, an integration of a spin-functionality in semiconductor devices, where the carrier concentrations are tunable and fully ballistic devices are possible, seems[38] the next step. Indeed, in the same era of time, Datta and Das proposed a new, heavily quoted device concept: an electronic analogue of the electro-optic modulator.[54] This device concept is based on spin injection from Fe-contacts in a ballistic high-electron-mobility transistor (HEMT)-like structure. The triangular confining potential of the two-dimensional electron gas (2DEG) creates an asymmetry in the semiconductor which results in an additional, direction dependent spin-splitting of the bands, similar to the DP-mechanism. This phenomenon - Structural Inversion Asymmetry (SIA), in contrast to Bulk Inversion Asymmetry (BIA) that leads to the DP-mechanism, has been predicted by Bychkov and Rashba.[55] This splitting of the bands results in an effective magnetic field, perpendicular to the propagation direction of the electrons, causing precession of the injected spins around the effective field. By voltage gating of the 2DEG this asymmetry can result in a change of the splitting and hence a change of the precession frequency. Using a second ferromagnetic electrode as an analyzer, the induced spin precession results in a modulation of the conductance. The splitting of the bands depends strongly on the propagation direction of the electrons and is averaged out over the entire Brillouin zone. Hence, a quasi-one-dimensional device is needed to obtain the largest effect. This device has been quoted numerous times as a new transistor, a spin transistor. However, for transistor operation, a significant gain is necessary and recent calculations have shown that the Datta-Das device does not perform well in terms of transistor characteristics.[56] Nevertheless, it has triggered a lot of interest and several groups have tried - up to now without much success - to fabricate such a device.

The first hurdle to take is to unambiguously show spin injection in semiconductors. The choice of the semiconductor material (InAs) seemed straightforward, at the InAs-surface the Fermi level pins in the conduction band, which leads to low-resistance ohmic contacts with metals. Furthermore, in the narrow-gap semiconductors such as InAs, the effects of SIA are predicted to be much larger than in GaAs. Therefore, the first attempts were made using ferromagnetic electrodes, typically $\text{Ni}_{0.8}\text{Fe}_{0.2}$ on an InAs inversion layer. The choice for ferromagnetic metals was made based on their high Curie temperature and their high intrinsic spin polarization. The aim was to demonstrate a difference in the device resistance for parallel and anti-parallel alignments of the electrode magnetizations (the

spin-valve effect). Several published reports indeed claim to have observed such a spin-valve effect, but the reported magnetoresistance was very small and subject to a lot of objections.[57; 58; 59; 60; 61; 62] The research of Monzon *et al* showed that the stray fields induced by the ferromagnetic electrodes generate a large local Hall effect (LHE), especially in the low-density two-dimensional electron systems.[63] This LHE strongly resembles the expected signal of the spin-injection-detection effect and most of the results could be explained likewise. Furthermore, Filip *et al* showed, using the before mentioned non-local technique, that they could not detect any magnetoresistance caused by the influence of electrical spin injection.[64]

The persistent lack of convincing results in these structures led to the conviction that there was a fundamental issue preventing an efficient spin injection in these structures.

The conductance mismatch.

Schmidt *et al* showed that in structures with ohmic contacts between ferromagnetic electrodes and semiconductors successful spin injection is problematic [65]. Although their calculations consider only diffusive transport in the regime of linear response, the results obtained in this study are quite general, at least qualitatively. The main problem is caused by the large difference in conductance between the ferromagnetic metal and the semiconductor. In a simple resistive picture, as shown in Fig. 2.10 where we consider the two spin channels not be intermixing in the semiconductors, the largest resistance in a ferromagnetic metal/semiconductor structure is provided by the semiconductor. This resistance is in a first approximation spin-independent, which means that it is impossible to drive a significant spin current through the semiconductor.

The use of highly polarized ferromagnetic materials could provide a solution for this problem since effectively one spin channel is eliminated. Schmidt *et al*, however, showed that a spin polarization of at least 98% is required to bypass the conductance mismatch, a challenging requirement. Matching the resistance of the ferromagnetic material to the semiconductor, by using e.g. ferromagnetic semiconductors as injector materials is an interesting route. One must note, however, that up to now the Curie temperatures of the established ferromagnetic semiconductors such as (Ga,Mn)As are still limited to below 200 K, which prohibits room temperature applications. Furthermore, the (III,Mn)V ferromagnetic semiconductors are p-type and holes have only very short spin lifetimes in semiconductors. This makes it difficult to use them for spin injection. A way to circumvent this problem is using interband tunneling as a tool to convert holes to electrons. This route will be explored in Chapter 6.

Finally, the ferromagnetic semiconductors with high Curie temperature,

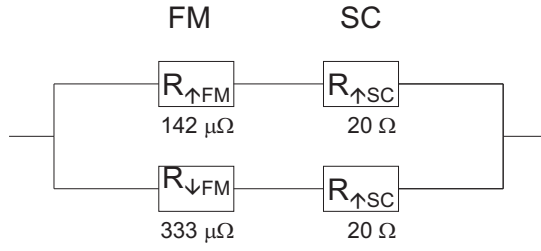


Figure 2.10: A simple resistive scheme of a ferromagnetic metal/semiconductor junction. The resistance associated with the spin channels in the metal and the semiconductor are realistic parameters for a ferromagnetic metal and a highly doped semiconductor with a spin-flip length exceeding one micrometer. It is clear that no significant spin current can flow in the semiconductor with this injection geometry.

are heavily doped, which means that also their resistance is not matched perfectly to the resistance of most normal semiconductors.

Filip *et al* already proposed that the introduction of a spin-dependent interface resistance that separates the ferromagnetic metal and the semiconductor may solve the problem. They proposed the use of a tunnel barrier between the ferromagnetic metal and the semiconductor. The current through the tunnel barrier depends on the density of states left and right of the tunnel barrier. Since the density of states in the ferromagnetic metal differs for spin-up and spin-down electrons, a tunnel barrier can provide a spin-dependent resistance. This idea has later been theoretically analyzed and approved by Rashba[66] and Fert and Jaffrès[67]. There are a few possibilities to create a tunnel barrier between a ferromagnetic metal and GaAs. A first one is using a scanning tunneling microscope (STM) with a ferromagnetic tip. In vacuum conditions, by applying a bias between the tip and the semiconductor, the tip can scan the bandgap and electron tunneling in the semiconductor conduction band can take place. Alvarado and Renaud showed, using a Ni tip, already in 1992 that spin polarized carriers could be created in bulk GaAs.[68] They monitored the polarization of the luminescence to estimate the spin-polarization of the injected carriers. A second possibility is using the native Schottky barrier between a metal and a semiconductor. Due to the difference in work functions or interface states induced Fermi level pinning, most metal-semiconductor combinations do exhibit a Schottky barrier.[69] Hence, an ohmic contact between a metal and a semiconductor is almost impossible, except in the case of InAs, where the Fermi level pinning occurs in the conduction band. Hence, electron injec-

tion itself requires the use of a tunnel barrier, such as the Schottky barrier. When this barrier is sufficiently thin, e.g. by tailoring the doping concentration in the semiconductor, tunneling can become the dominant charge transport mechanism in the Schottky diode. Finally, an artificial tunnel barrier can be created by inserting an insulating material, such as AlO_x , between the metal and the semiconductor. This has the advantage that the metal and the semiconductor are physically separated. Hence, problems with chemical stability of the metal/semiconductor interface are less of an issue here.

2.4 Tunneling

The realization of efficient spin injection in semiconductors at room temperature requires the use of tunnel barriers that act as a spin-dependent interface resistance between the ferromagnetic material and the metal. In this section three types of tunneling into semiconductors will be reviewed. The first two deal with tunneling from (ferromagnetic) metals into semiconductors, namely by tunneling through the Schottky barrier that arises at the metal/semiconductor interface and through an oxide tunnel barrier. These two mechanisms will be used experimentally, to achieve spin injection in GaAs. Finally, a third part will focus on interband tunneling between the valence and the conduction band of a semiconductor. This part is relevant for tunneling of spin-polarized electrons from a p-type ferromagnetic semiconductor. As holes have only short spin-lifetimes in semiconductors, an efficient conversion between holes and electrons is necessary and can be provided by the interband tunneling mechanism.

2.4.1 Tunneling in metal/semiconductor structures

Tunneling from electrons from a metal into a semiconductor is a transport process between two very different electronic systems. The large Fermi energy in the metal and the corresponding large Fermi wave vector are not matched well to a semiconductor, exhibiting a low Fermi energy and a small Fermi wave vector. Correspondingly, the tunneling process, where generally a conservation of the in-plane quasi-momentum (perpendicular to the current) is assumed, is dominated by the part of the metal Brillouin zone, with a small in-plane momentum (perpendicular to the current) and a large out-of-plane momentum (parallel to the current). This is illustrated in Fig. 2.11, where is shown that for a very simplified metal band structure, indeed only a limited number of states can participate in the tunneling process. Furthermore, electrons that travel through the tunnel barrier and have a relatively large in-plane quasi-momentum have a longer trajectory

in the tunnel barrier and hence exhibit a larger damping, which effectively reduces their tunnel probability. It is clear that the spin polarization, as determined by integrating the total density of states, does not necessarily correspond to the spin polarization of the tunneling electrons. E.g. Prins *et al* tried to demonstrate electrical spin detection of optically injected spins in GaAs.[70] Since the optically injected spins are oriented out-of-plane, an out-of-plane magnetized ferromagnetic metal was chosen for the experiment, namely epitaxially grown MnAl. As a tunnel barrier they chose AlAs. They didn't succeed in measuring any spin-related signal and attributed it to the tunneling process. Ab initio band structure calculations indicated that the spin-polarized electrons mainly occupy the states with an in-plane momentum, while electrons travelling perpendicular to the plane (i.e. parallel to the magnetization) only exhibit a very low spin polarization. Since tunneling mainly involves electrons with a considerable out-of-plane momentum, no signal was expected. In any epitaxial structure, the details of the band structure will be very important to understand the absolute value of the injected spin polarization and its bias dependence.

The details of the damping in the forbidden gap are very important in order to get a good description of the tunneling process. Therefore, a good knowledge of the band structure of the semiconductor and/or the insulator is necessary. The conduction and the valence band states are characterized by their energy and their momentum (k). Within the band gap, the momentum (k) becomes imaginary. The conduction and the valence bands are connected through the band gap by a trajectory in the (E - k)-space. This is depicted in Fig. 2.12, where a simplified (E - k^2)-diagram is shown for a semiconductor. Especially when different bands contribute to the tunneling current, this becomes important in order to understand the tunneling process.

These arguments account mainly for the case of epitaxial systems, where the assumption of conservation of the in-plane wavevector is justified. In the case of polycrystalline metals and especially amorphous tunnel barriers, it is less clear what happens. The in-plane periodicity of the lattice is broken, and hence the in-plane quasi-momentum may not be conserved in the tunneling process. Also in the case of polycrystalline materials, the details of the band-structure may be important, as long as a strong texturing of the crystal orientation is present, as indicated in the example of the MgO-based magnetic tunnel junctions.

Tunneling in metal/semiconductor structures

A metal contact to a semiconductor generally leads to the formation of a Schottky barrier. The misalignment of the Fermi level in the semiconductor and the metal work function, or the misalignment of the bulk semiconduc-

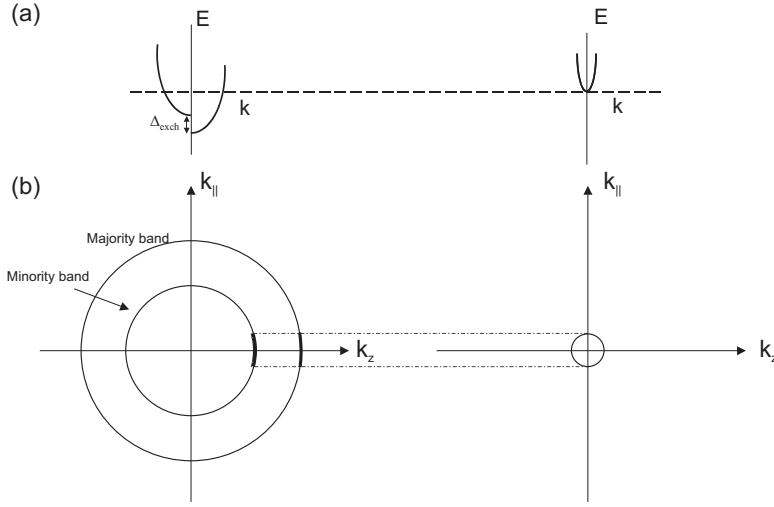


Figure 2.11: (a) The energy (E) vs quasi-momentum (k) schematic diagrams for a ferromagnetic metal and an n-type semiconductor. The bands of the ferromagnetic metal are split by the exchange interaction. (b) The cross-section of the E - k -dispersion diagrams at the Fermi energy for both the ferromagnetic metal and the semiconductor. We assume parabolic bands and plot the parallel quasi-momentum ($k_{\parallel}$) as function of the perpendicular quasi-momentum (k_z). It is clear that the states in the ferromagnet that can participate in the tunneling process are very limited.

tor Fermi level and the surface Fermi level (which is generally different due to midgap Fermi level pinning by surface states) creates a charge transfer between the semiconductor and the metal or the surface states. Since the high carrier density in the metal screens the electric fields, the potential difference is accommodated by band bending in the semiconductor. Hence, the Schottky barrier is formed by the space charge region in the semiconductor. When the semiconductor is heavily (n-type) doped, the bulk Fermi level lies within the conduction band, as can be seen on Fig. 2.13(b). Furthermore, since the width of the depletion region is inversely proportional to $\sqrt{N_D}$, the square root of the donor concentration, in the case of high doping the barrier is sufficiently thin to allow tunneling between the semiconductor and the metal. The tunnel probability is determined by the height and the thickness of the tunnel barrier, and the behavior of the (E - k)-relation within the gap. Since the Schottky barrier has a more or less triangular shape, high energy electrons see a lower tunnel barrier height and a reduced thickness, which

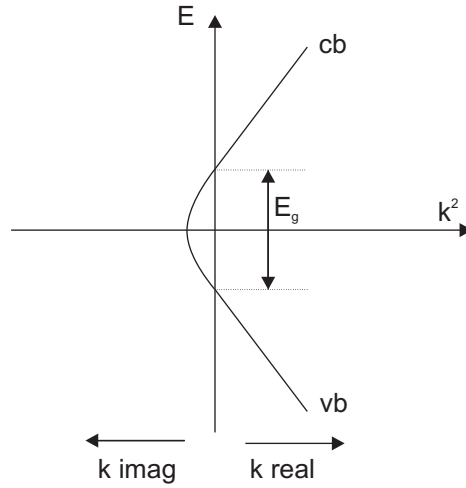


Figure 2.12: The simplified complex band diagram of a semiconductor. The details of the band structure in the imaginary k -plane determine to a great extent the tunnel current.

causes the tunnel probability to be strongly energy dependent. The tunnel probability for a triangular tunnel barrier can be written as follows:[71]

$$T = \exp \left(-\frac{4}{3} \frac{\sqrt{2qm^*}}{\hbar} \sqrt{E_b} d \right), \quad (2.17)$$

with q the electron charge, m^* the effective mass in the tunnel barrier, $\hbar$ Planck's constant, E_b the tunnel barrier height and d the tunnel barrier thickness. To get an estimate of the energy resolved tunnel current through such a barrier, the tunnel probability should be multiplied by the energy distribution of the electron occupation, the Fermi-Dirac distribution:

$$F(E) = \frac{1}{1 + \exp \left(\frac{E - E_F}{kT} \right)}, \quad (2.18)$$

with E_F the Fermi energy, k the Boltzmann constant and T the temperature. In Fig. 2.13(b) the tunnel probability is multiplied with the Fermi-Dirac distribution as function of the energy in the case of a Schottky barrier (simplified by a triangular tunnel barrier) for two values of the donor concentration, namely $1 \times 10^{19} \text{ cm}^{-3}$ (left) and $1 \times 10^{18} \text{ cm}^{-3}$ at room temperature. It is clear that for a lower doping concentration, and hence a thicker tunnel barrier the main part of the tunnel current originates from high energy electrons in the metal layer. This means that the spin polarization at the Fermi

energy, which is generally used as a figure of merit for ferromagnetic materials, is not important in this case. Also in the case of electrons tunneling into the ferromagnetic metal layer, certainly under bias, they will tunnel into higher energy states, with a different characteristic spin polarization.

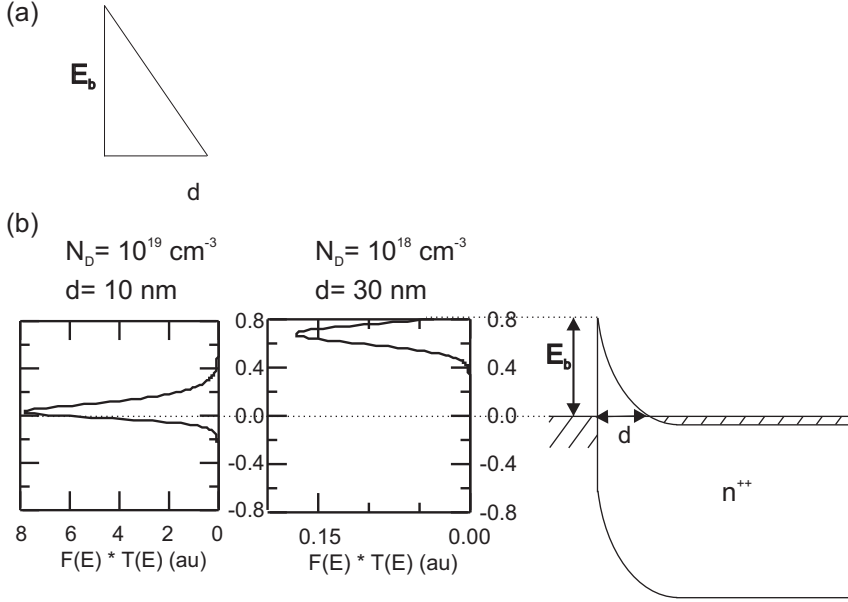


Figure 2.13: A simplified picture of the energy dependence of the injected electrons. (a) The triangular tunnel barrier used for calculating the tunnel probability. (b) The Schottky barrier (right) and the product of the Fermi-Dirac distribution with the tunnel probability as function of the energy for two different doping levels in the semiconductor. It is clear that for a lower doping level, the average energy of the injected electrons increases drastically.

Since in Schottky diodes the tunnel barrier is formed by the depleted semiconductor, which is crystalline and generally of high quality, band-structure effects are expected to play a role in the tunneling process. Indeed, Schottky barrier tunnel diodes have been employed as tools to determine the $(E-k)$ -relation within the semiconductor band gap. Conley and Mahan [72] have used tunneling spectroscopy from Schottky barriers on GaAs to measure the $(E-k)$ -relation and the density of states in GaAs, for both p- and n-type GaAs.

Tunneling in metal/oxide/semiconductor structures

Metal-oxide-semiconductor structures have been heavily investigated in the past due to the important role they play in metal-oxide-semiconductor field effect transistors (MOSFETS). In the process of continuous scaling of the transistor dimensions, the (silicon) oxide thickness has reached the regime where tunneling starts to play an important role. Hence, the tunneling processes in these structures are well documented.

Due to the rectangular shape of the tunnel barrier (or more generally trapezium shape) and the high barrier height, the tunnel current is dominated by electrons at the metal Fermi level, unlike in the case of the Schottky barriers. Oxide barriers are often amorphous, which generates a less defined (E-k)-relation.

2.4.2 Interband tunneling in semiconductor heterostructures

Esaki was the first to investigate interband tunneling, when he and his coworkers discovered a negative differential resistance in heavily doped Ge $p-n$ -diodes that were forward biased.[73] Furthermore, the current at reverse bias exceeded the current at forward bias, contrary to the expected rectifying behaviour of the diode. They discovered that interband tunneling is the basic physical mechanism behind this effect. The band line-up for a heavily doped $p-n$ -diode is sketched in Fig. 2.14

Both in forward and in reverse bias electrons tunnel between the conduction and the valence band. In forward mode, however, electrons tunnel into the empty states in the valence band of the p -layer and when the applied voltage exceeds roughly the Fermi energy in the p -layer, no electronic states are available in the p -layer due to the energy gap and the tunnel current collapses.

The interband tunneling heterostructure is typically a fully epitaxial structure. Furthermore, the electrons follow the full trajectory from valence band to conduction band (or vice versa), as depicted in Fig. 2.12. Hence, the details of the (E-k)-relation are very important to fully understand the tunneling process.

2.5 Conclusions

In this Chapter we discussed the research field in which this work is embedded. Both elementary physics and a historical overview of research in spin-dependent electronics in metals and semiconductors are presented. Early experiments on spin injection in metals and semiconductors are discussed,

together with the conductance mismatch, a fundamental obstacle for spin injection in semiconductors. As a solution for this problem, tunnel contacts have been proposed before and will be used in this thesis. Different types of tunnel contacts will be used and the tunnel process has been discussed in this Chapter.

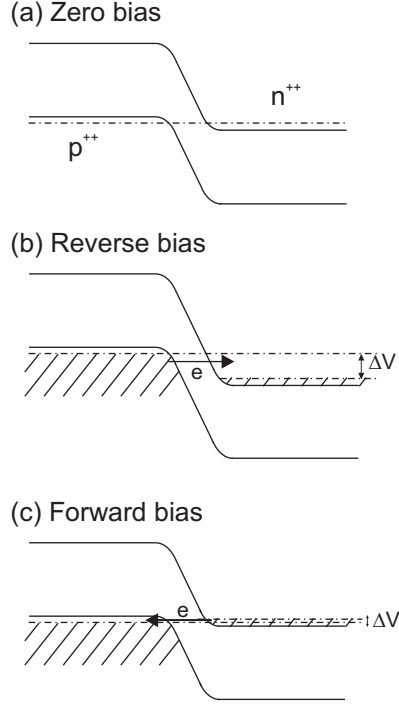


Figure 2.14: (a) The band line-up of the diode at zero bias. Both the p -layer and the n -layer of the device are degenerate. (b) Upon applying a reverse bias, valence band electrons from the p -layer tunnel through the forbidden gap into the conduction band of the n -layer, resulting in a large reverse current. (c) At low values of the applied forward bias electrons can tunnel from the n -layer into the empty states in the valence band of the p -layer. Increasing the bias voltage blocks this effect, resulting in a negative differential resistance. At higher values of the forward bias voltage, the regular $p - n$ -diode current characteristics dominate.

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Chapter 3

Spin detection in semiconductors: spin light-emitting-diodes

3.1 Introduction.

The detection of spin-polarized carriers in semiconductors by electrical means has been shown to be far from straightforward. Measuring the difference in resistance in an injection-detection device between the cases where the injector and detector electrodes exhibit a parallel and an anti-parallel magnetization is subject to quite some side-effects, such as the local Hall effect and anisotropic magnetoresistance of the contacts. Furthermore, since the injection and the detection efficiencies are both unknown parameters, the genuine injected spin polarization is hard to extract from such experiments. An alternative way is offered by the well-known optical selection rules in III-V semiconductors, which have been introduced in Chapter 2. The recombination of spin-polarized electrons with unpolarized holes results in the emission of circularly polarized light, the circular polarization being proportional to the spin polarization in the direction of observation. This technique can hence, in principle, result in a direct measure of the injected spin polarization. The fabrication of devices with this kind of functionality can be deduced directly from the wide-spread knowledge on light-emitting-diodes, typically p-i-n-diodes that emit light in forward bias. By creating a magnetic contact that injects electrons in such diodes, a steady-state spin polarization can be created and detected. This approach has been suggested and patented already in 1993 by Jonker.[1]

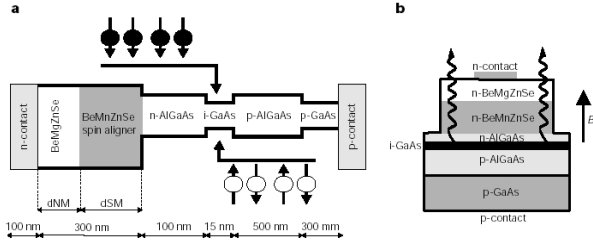


Figure 3.1: (a) The schematic band-structure of the surface emitting spin-LED used by Fiederling *et al.* (b) The device in cross-section.

The first successful implementation of the spin-light-emitting-diode concept (spin-LED) has been shown by Fiederling *et al.*[2] As indicated on Fig. 3.1, they used the dilute magnetic semiconductor (DMS) $\text{Be}_x\text{Mn}_y\text{Zn}_{1-x-y}\text{Se}$ as spin aligner between the electrical contact and the LED. By applying a relatively large magnetic field parallel to the growth direction, the giant Zeeman splitting creates a full polarization of the carriers in the DMS-layer. If the thickness of this layer is significantly larger than the spin-diffusion length in this material, the electron spins will be fully polarized when entering the AlGaAs transport layer. Using this device, they were able to achieve an injected spin polarization of 90%, determined from the circular polarization of the luminescence. Similar experiments, with similar results have been achieved simultaneously by Jonker *et al.*[4] At the same time, Ohno *et al* published results on electrical spin injection of holes from (Ga,Mn)As into an (In,Ga)As quantum well.[3] The circular polarization of the light emitted along the edge of the sample (perpendicular to the growth direction) was measured to be about 1%. Although the circular polarization is correlated with the magnetization state of the (Ga,Mn)As injector, the results are difficult to interpret due to the different selection rules for edge-emission (side emission) and surface emission using quantum wells.[5]

At the start of this research project, some groups already reported electrical spin injection from a ferromagnetic metal into a spin-LED. Zhu *et al*[6] and Hanbicki *et al*[7] reported spin injection from an epitaxial Fe contact layer into a GaAs/InGaAs spin-LED and an AlGaAs/GaAs spin-LED, respectively, utilizing the Schottky barrier as tunnel barrier. At low temperature, they reported 2% at 80 K and 13% at 5 K injected spin polarization, respectively. In the report of Zhu *et al* the measured steady-state spin polarization stayed 2% at room temperature, while in the report of Hanbicki *et al* the injected spin polarization steadily decreased, to reach 4% at 240 K. In Imec Motsnyi *et al* used a thin AlO_x insulating layer as a

tunnel barrier for the injection of spin polarized electrons from a sputtered CoFe top contact into an AlGaAs/GaAs spin-LED.[8] They demonstrated a steady-state spin polarization of 8% at 80 K.

3.2 Spin-manipulation: the oblique Hanle effect.

As has been mentioned before, the degree of circular polarization in a spin-LED is proportional to the spin polarization in the direction of observation. In typical ferromagnetic thin films, the shape anisotropy forces the magnetization in the plane. The light emitted in a surface emitting diode is collected in a direction perpendicular to the easy axis of the magnetization. Hence, no circular polarization is expected. This could be solved by collecting the light parallel to the magnetization, i.e. measuring the light emitted from the edge (side emission). This method, however, suffers from several background effects, such as multiple reflections at the metal/semiconductor interface during the propagation of the light in the semiconductor that can change the state of the circular polarization, reabsorption in the semiconductor, etc. The cleanest detection method is measuring in a surface-emitting mode. In this case the spins should be manipulated to acquire a non-zero perpendicular component, which is reflected in the circular polarization of the light.

There are two ways of manipulating the spins: Firstly, by manipulating the spins in the ferromagnetic material by applying a strong external field parallel to the growth direction that pulls the magnetization out of plane. Secondly by manipulating the spins in the semiconductor by applying a relatively small external magnetic field that is non-collinear with the injected spins. Under certain conditions the induced precession will create a non-zero perpendicular component of the average electron spin. The first geometry (the Faraday geometry) has been applied by a number of groups. It has the advantage of simplicity. When the magnetization is saturated along the magnetic field, the degree of circular polarization is simply one half of the degree of spin polarization in the case of recombination in a bulk semiconductor, or equals the degree of spin polarization in case of strain or quantization. It has the disadvantage that the field needed for the transition from in-plane to out-of-plane magnetization is typically larger than 2 T. In semiconductors with a large g-factor, such as InAs based compounds, this may lead to a large thermal spin polarization at low temperatures, both in the conduction band and in the valence band. This may complicate the interpretation of the experiments.[9] The second, precessional method, has the advantage that only a small external field is necessary and that information about the measured circular polarization contains information

about the spin dynamics in the semiconductor, as will be shown in the next paragraphs.

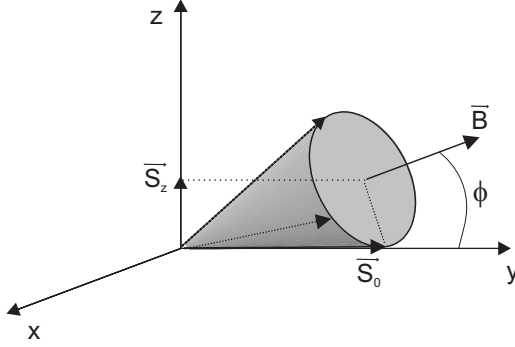


Figure 3.2: Schematically is shown that the precession induced by the application of an oblique magnetic field leads to a perpendicular spin-component.

Motsnyi *et al* already developed a technique based on this principle. Their technique has been inspired by the oblique Hanle effect, a technique originally developed for the visualization of a nuclear field in optical pumping experiments, as already noted in Chapter 2. They applied an oblique magnetic field, oblique with respect to the plane, which caused spin precession in the semiconductor. As schematically shown in Fig. 3.2 this leads to a perpendicular spin-component, observable through the circular polarization of the emitted light.

The dynamics of the injected spins are governed by the Bloch equation, that is described in eq. 2.6. We can solve this equation readily for the geometry that is shown in Fig. 3.3. We consider a steady-state situation, hence $d\mathbf{S}/dt = 0$. In this situation S_Z exhibits the following dependence on the external magnetic field:

$$S_Z(B) = S_{0x} \cdot \frac{T_s}{\tau} \cdot \frac{(\Omega T_s)^2 \cdot 1/2}{1 + (\Omega T_s)^2} \quad (3.1)$$

The perpendicular spin component equals zero at zero field and increases upon the application of the oblique magnetic field. At high magnetic field, when the injected spins have undergone several precession cycles before recombining, S_Z saturates. The saturation value depends on the angle between the magnetic field and the plane and can be maximized by taking this angle equal to 45° , as indicated in Fig. 3.3

Using the selection rules for bulk GaAs, as depicted in eq. 2.4, the following relation can be derived between the perpendicular spin component

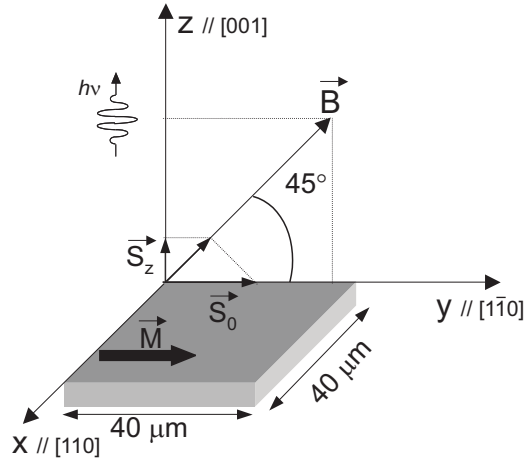


Figure 3.3: A schematic picture of the measurement geometry. The magnetic field is applied at an angle of 45° with respect to the plane and the magnetization. The magnetization is in plane, along one of the two in-plane crystal axes $[110]$ or $[1\bar{1}0]$, while the light is emitted along the $[001]$ -axis. Due to the precession, the average spin $\mathbf{S}$ acquires a non-zero perpendicular component $\mathbf{S}_z$.

and the circular polarization of the emitted light:

$$P_{circ} = S_z \quad (3.2)$$

Hence, a measure of $P_{circ}(B)$ yields the steady state spin polarization in the semiconductor active region. The steady state spin polarization is related to the injected spin polarization Π_{inj} by a factor that takes into account the loss of spin-polarization between the injection and the recombination of the carriers: $\Pi_{steadystate} = \Pi_{inj} \frac{T_S}{\tau}$.

3.2.1 Expected behaviour of the degree of circular polarization in a spin-LED.

The behaviour of the circular polarization of the light emitted in a spin-LED is not only governed by the Bloch equations, as explained in the previous paragraph, but also by the behaviour of the magnetization of the ferromagnetic injector. In Fig. 3.4 the theoretical behaviour of the degree of circular polarization is shown as function of the external magnetic field for two different ferromagnetic injectors, one with a small (a) and one with a large (c)

coercivity. We assume at this stage that the magnetization stays in-plane upon the application of an oblique external magnetic field.

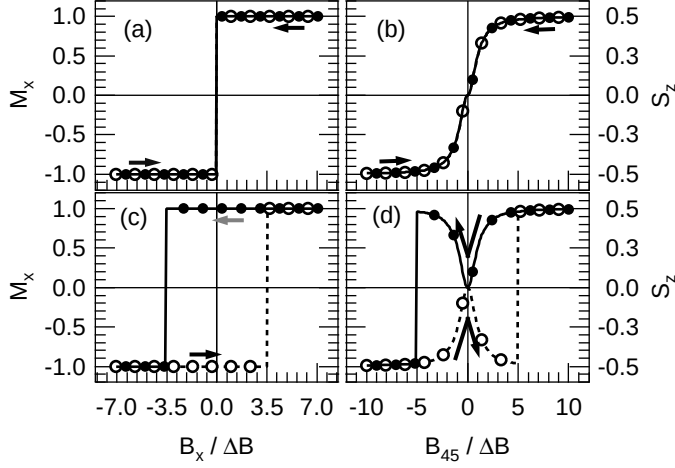


Figure 3.4: (a) The in-plane magnetization characteristics of a ferromagnetic material with a small coercivity and (b) the average perpendicular spin component in the semiconductor versus the magnetic field, as expected for spin injection from a ferromagnetic material with magnetic characteristics as depicted in (a). (c) The in-plane magnetization characteristics for ferromagnetic material with a large coercivity and (d) the average perpendicular spin component in the semiconductor versus the magnetic field, as expected for spin injection from a ferromagnetic material with magnetic characteristics as depicted in (c).

3.3 Calibration experiments and side effects.

The use of a GaAs spin-detector, embedded in a spin-LED results in the determination of the steady-state spin polarization in the active region of the semiconductor. This yields, however, only a first approximation of the injected spin polarization, since part of that is lost due to spin scattering in the transport to the active region, in the thermalization process to the bottom of the conduction band and during the stay of the electrons at the bottom of the conduction band before recombining with holes. Furthermore the oblique Hanle effect analysis is disturbed by some side effects that spuriously increase the degree of circular polarization.

Calibration experiments. The efficiency of the spin relaxation process can be determined independently by an optical pumping experiment. Indeed, by irradiating the sample with circularly polarized light, spin polarized carriers are created with an initial spin polarization $P = 0.5$. Measuring the Hanle depolarization curve under continuous optical pumping yields a direct measure of both T_S and τ .

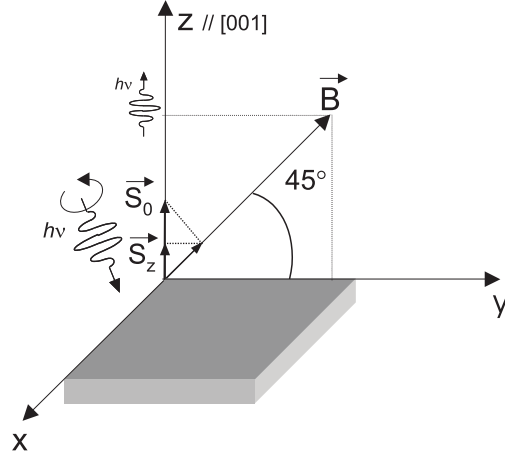


Figure 3.5: A schematic picture of the measurement geometry. The magnetic field is applied in an angle of 45° with respect to the plane. The light impinges on the semiconductor top surface, and due to the large diffraction index of GaAs, enters GaAs quasi-parallel to the growth direction. This causes the created electron spins to be oriented along the growth direction. The application of an oblique magnetic field causes the electron spins to precess around the field, destroying the component of the average spin perpendicular to the field.

The measurement geometry of such an optical pumping experiment is depicted in Fig. 3.5. The created electron spins are oriented along the light beam, which is always along the growth direction for light impinging the top surface due to the large diffraction index of GaAs. Upon the application of an oblique magnetic field the injected spins start to precess around the field and the component of the average spin perpendicular to the external field is destroyed. This causes S_Z to decrease with increasing magnetic field down to its saturation value, which occurs when the injected spins have undergone several precession cycles before recombining. The dependence of $P_{circ} = S_Z$ on the external field can be described as follows:

$$P_{circ} = S_{0z} \cdot \frac{T_s}{\tau} \cdot \frac{1 + (\Omega T_s)^2 \cdot \cos^2 \phi}{1 + (\Omega T_s)^2} \quad (3.3)$$

Measuring this dependence yields the experimental values of T_s and τ .

Side effects. The measurement process involves however some side effects: The application of an external magnetic field will create a *thermal spin polarization in the semiconductor* (the Zeeman-effect), giving rise to a spin-signal in the degree of circular polarization, not originating from spin injection. Furthermore the oblique magnetic field will tilt the magnetization of the thin magnetic film out of plane. The field induced out-of-plane component of the magnetization alters the experimental detection method in two ways. Firstly, the out-of-plane component of the magnetization causes a helicity dependent absorption for light traversing the ferromagnetic thin film, *magnetic circular dichroism* (MCD). This creates an additional source of circular polarization, independent of the spin-polarization of the recombining electrons. These effects can be measured independently from the spin-injection experiment by an optical pumping experiment. Exciting the semiconductor by linearly polarized light at an excitation energy of 1.96 eV (excitation energy of a He-Ne laser, which causes also transitions from the split-off band), causes the creation of electron-hole pairs without a net-spin polarization. The external magnetic field can cause them to acquire some degree of spin-polarization before recombining. Upon recombination in the GaAs active region, they emit light - with a degree of circular polarization corresponding to the thermal spin polarization, which acquires some additional circular polarization after the traversal of the ferromagnetic thin film. The obtained field-dependence of the circular polarization can be subtracted afterwards from the results obtained by electrical spin injection, resulting in the circular polarization signal obtained by spin-injection only.

Apart from a direct influence on the degree of circular polarization, the tilting of the magnetization changes slightly the analysis employed to obtain the degree of injected spin polarization. The injected average electron spin, previously assumed to be perfectly in-plane, has a *perpendicular component*, depending on the external magnetic field. This average injected electron spin can be described as follows: $\mathbf{S}_0(0, S_0 \cdot \cos(\theta), S_0 \cdot \sin(\theta))$ with θ the angle between the magnetization and the plane. We can adapt the equation to describe the perpendicular component of the average electron spin and hence the degree of circular polarization of the emitted light as follows:

$$P_{circ}(B) = (S_0 \cdot \cos \theta) \frac{T_s}{\tau} \cdot \frac{(\Omega T_s)^2 \cdot 1/2}{1 + (\Omega T_s)^2} + (S_0 \cdot \sin \theta) \frac{T_s}{\tau} \cdot \frac{1 + (\Omega T_s)^2 \cdot 1/2}{1 + (\Omega T_s)^2} \quad (3.4)$$

The equation for P_{circ} now consists of two terms, the first one describing the influence of the in-plane component of the injected electron spin and the second one describing the out-of-plane component of the injected electron spin. The angle between the magnetization and the plane depends on the external field and the total magnetization. The density of the shape anisotropy energy, is given by $U_d = -\mu_0 \cdot (\mathbf{M} \cdot \mathbf{H}_d)/2$, with $\mathbf{M}$ the magnetization and $\mathbf{H}_d = \mathbf{M}_z$ the demagnetization field, with $\mathbf{M}_z$ the perpendicular component of the magnetization. The total energy density of the ferromagnetic thin film in an external magnetic field H_{ext} is given by:

$$\begin{aligned} U &= -\mu_0 \cdot (\mathbf{M} \cdot \mathbf{H}_{ext}) - \frac{\mu_0 \cdot (\mathbf{M} \cdot \mathbf{H}_d)}{2} \\ &= -\mu_0 \cdot (M \cdot H_{ext}) \cdot \sin(\theta) + \frac{\mu_0 \cdot M^2}{2} \cdot \sin^2 \theta \end{aligned} \quad (3.5)$$

Minimizing this equation results in the following relation between the angle θ and the applied field B :

$$\frac{B}{\mu_0 \cdot M} = \frac{\sin \theta \cdot \cos \theta}{\sqrt{2}/2 \cdot \cos \theta - \sqrt{2}/2 \cdot \sin \theta} \quad (3.6)$$

Hence, if we know the magnetization of the ferromagnetic material, we can fully take into account the effect of the tilting of the magnetization.

3.4 The design of spin-LEDs.

The design of a spin-LED involves in the first place the design of a light-emitting diode. The electrons and the holes must be brought close to each other to allow the necessary recombination. Furthermore, the electron-hole recombination time should be significantly shorter than the spin-relaxation time to get a fairly good measure of the injected spin polarization. The transport of the injected electron spins towards the recombination region is also an important issue.

In Fig. 3.6 is schematically shown how a spin-LED is designed. Under bias spin-polarized electrons should be able to tunnel from a ferromagnetic contact through a tunnel barrier in the conduction band of AlGaAs. The spin-transport region should be lightly n-type doped or undoped and allow transport of the spins without being scattered or diluted. In a highly doped n-type region the DP-mechanism leads to efficient spin-relaxation. The holes are supplied by the p-type substrate and are collected, together with the electrons in the GaAs recombination layer. The AlGaAs-barriers allow confinement of the electrons and the holes, such that efficient recombination can take place. If the thickness of this layer is made sufficiently thin,

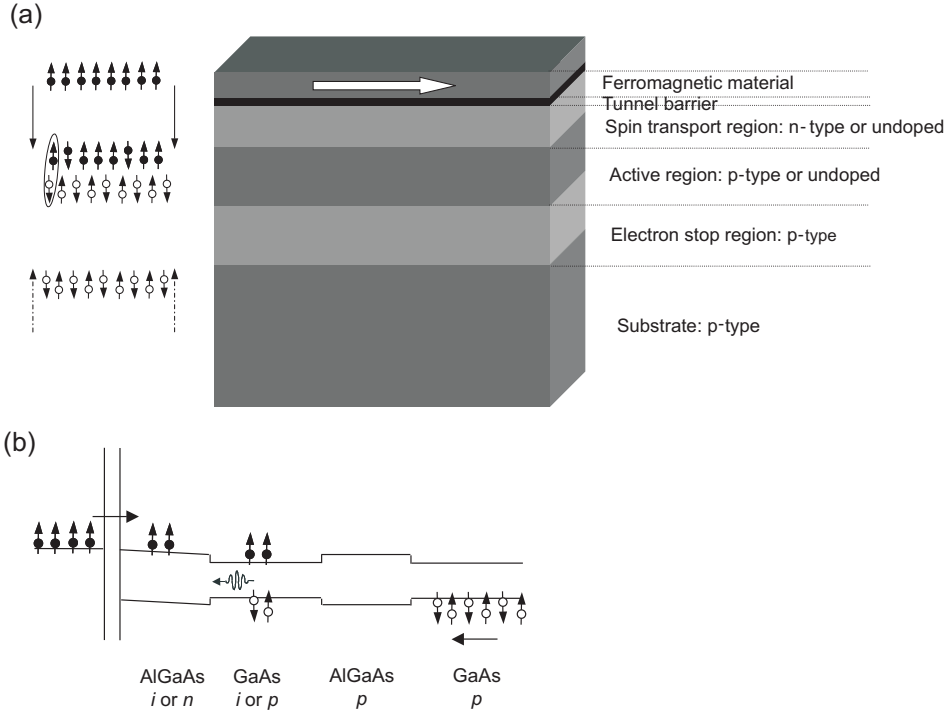


Figure 3.6: (a) A schematic cross section of a generic spin-LED. (b) The band structure under bias of a generic AlGaAs/GaAs-based spin-LED.

quantization of the energy levels takes place, such that the heavy and the light hole bands split, resulting in changing selection rules. In this work, the thickness of the active region will be chosen such that bulk selection rules can be employed. Therefore a thickness of 100 nm will be chosen. The active region can be p-type doped or undoped. A p-type doping shortens the electron-hole recombination time, which enhances the spin-detector properties. The p-type doping, however, shortens also the spin-relaxation time, through the BAP-spin relaxation mechanism. Hence, both types of doping will be used, depending on the specific structure.

3.4.1 Medici simulations

In order to accurately engineer both the bands (through the Al-concentration) and the band bending (through the doping levels) of the semiconductor devices in this thesis, we perform self-consistent simulations using Medici. This

semiconductor simulation tool allows self-consistent calculations of semiconductor heterostructures taking into account Schottky barrier tunneling, direct tunneling through oxides, band-to-band tunneling, recombination and impact ionization. Also interface trap states can be included to simulate Fermi-level pinning, which is necessary in the case of GaAs. Special care has been taken that the Fermi level pinning of the GaAs surface is well reproduced by the simulations. Otherwise, reliable results are excluded. In the case of high doping ($> 10^{19}\text{cm}^{-3}$) a complete ionization of the impurities is assumed.[10] By these (spin-independent) simulations a detailed understanding of the charge distribution, band bending, electron and hole currents and their bias dependence in the entire device can be achieved.

3.5 The considered spin-LED-structures.

In this work, three types of tunnel injectors have been used to realize spin injection in GaAs. Aluminum oxide tunnel barriers have been employed to realize spin injection from a sputtered CoFe contact, the native Schottky barrier has been used to realize spin injection from an epitaxially grown MnSb contact metal and electron spin injection has been achieved from the p-type ferromagnetic semiconductor (Ga,Mn)As by interband tunnelling.

3.6 Spin-LED processing.

After the growth of the complete layer structure, the wafers have to be processed into light emitting devices, which can be appropriately biased. The processing scheme is depicted in Fig. 3.7. Depending on the material used for the ferromagnetic thin film, the metal injectors are patterned by a dry or a wet etch into mesas with different sizes, yielding from $20 \times 20\mu\text{m}$ to $100 \times 100\mu\text{m}$. After the etching procedure, an insulating layer of SiO_x is deposited by means of lift-off, which covers the AlGaAs/GaAs-wafer and part of the metal electrodes. In order to create the top contacts to the injector electrode, Au is deposited by means of lift-off, leaving an optical window through which the light can be emitted. Finally the backside of the wafer is metallised by Au to provide the back-contact to the p-GaAs wafer.

3.7 Measuring spin-LEDs.

The setup, used for the measurement of the circular polarization of the luminescence in the spin-LEDs, is depicted in Fig. 3.8. The sample is mounted in a cold-finger cryostat, which can be cooled by either liquid Helium or Nitrogen. The cryostat is positioned between the poles of an electromagnet, which

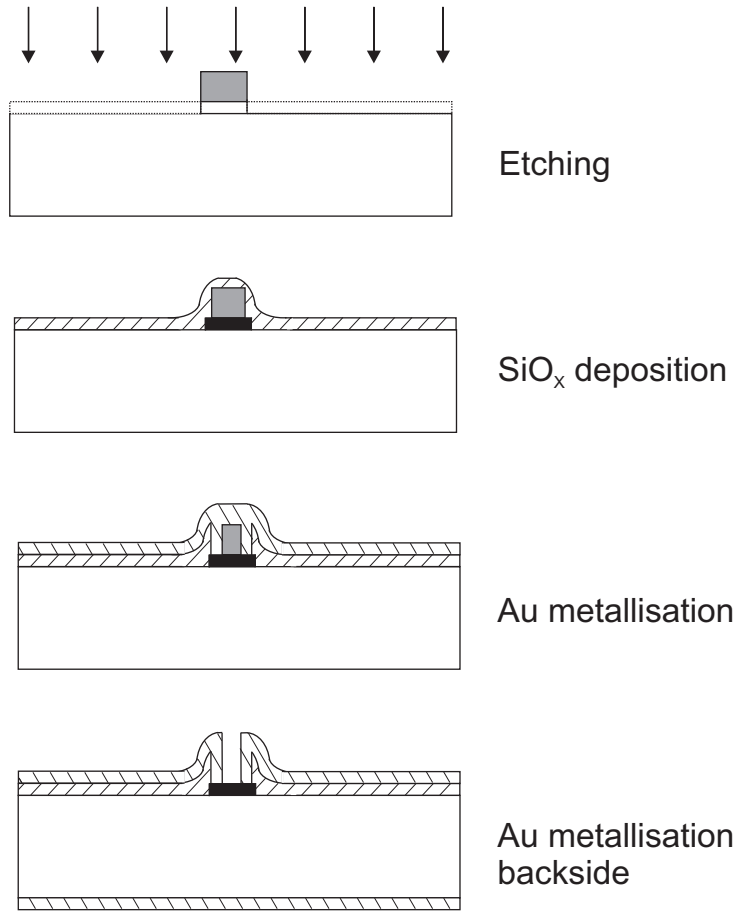


Figure 3.7: The processing of the spin-LEDs.

can generate a maximum magnetic field of 0.56 T. In the present measurement configuration, the electromagnet supplies an oblique magnetic field, and allows measurements following the principles of the oblique Hanle effect. A first lens, close to the optical window of the cryostat provides beam collimation and allows the emitted light to be transmitted through a polarization modulator and an analyzer, while a second lens focuses the beam either on an optical fiber, which is coupled into a detector, either on a monochromator and an attached detector, which allows spectral resolution. The employed polarization modulator is either a continuously rotating quarter wave plate, generating a quarter wave retardation at about 700 Hz,

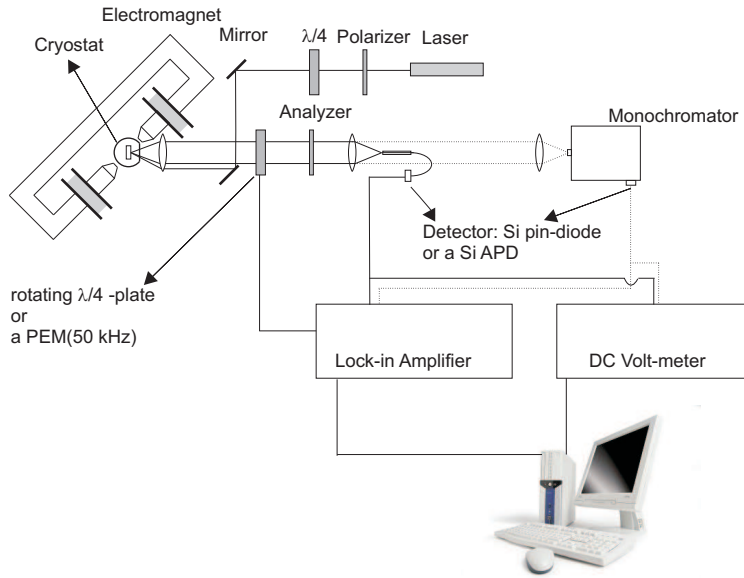


Figure 3.8: The used setup for the spin-LED measurements.

or a photo-elastic modulator (PEM), generating a quarter-wave retardation at 50 kHz. Both schemes essentially perform similarly: circularly polarized light is converted through the quarter-wave retardation into linearly polarized light, with mutually perpendicular orientations for σ^+ and σ^- polarizations, which is detected by the analyzer. Modulating between positive and negative retardation results in a modulation of the intensity of the light at the modulation frequency with an amplitude of $I^+ - I^-$. Using a lock-in amplifier, with the reference frequency from the modulator, allows to measure this value, while a DC-voltage meter allows to measure the total intensity, which yields the value for the circular polarization. The different modulation frequencies for the two approaches, result in different needs for the used detector. Hence, for the rotating quarter-wave plate, a very sensitive, Si pin-photodiode with a cut-off frequency of 1 KHz is used, while for the PEM a Si avalanche photo-diode (APD) with a cut-off frequency of 100 KHz is used.

The setup also allows to perform photoluminescence experiments for calibration experiments, such as the measurement of the MCD-effect as function of the applied magnetic field, and the determination of the spin and the electron lifetimes. The MCD-effect can be measured by exciting electron-hole pairs in the sample using linearly polarized light from the

combination of a He-Ne laser, with a 1.96 eV excitation energy and a polarizer, and measuring the circular polarization of the light. The spin and electron lifetimes can be determined by measuring the Hanle depolarization curve of the photoluminescence. In this case the combination of an AlGaAs-semiconductor laser, with a 1.56 eV excitation energy, a linear polarizer and a quarter-wave plate are used to generate spin-polarized electron-hole pairs in the semiconductor.

3.8 Conclusions

In this Chapter an optical technique to measure the polarization of injected carriers has been presented. We have chosen spin-LEDs as reliable spin detector systems. We have discussed the possible methods to analyze the polarization of the light and argued why we chose for the oblique Hanle effect, a technique based on the precession of electron spins in an oblique magnetic field. Furthermore, the different background effects that can interfere with the measurement have been discussed and calibration experiments have been proposed. Finally we presented the measurement setup used to experimentally determine the spin polarization of the injected carriers in the spin-LEDs.

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Chapter 4

Spin injection in an MIS-spin LED

4.1 Introduction

The realization of electrical spin injection by means of tunnel injection from a ferromagnetic metallic contact remains a challenging task. To circumvent the conductance mismatch, as pointed out in Chapter 2, the introduction of a tunnel barrier between the metal contact and the semiconductor, which acts as a large spin-dependent resistance is necessary. Such a resistance can be provided by a tunnel barrier as supported by recent calculations by Rashba and Fert.[1] Fert also calculated that the tunnel barrier resistance must be matched to the semiconductor channel resistance in order to result in a maximum magnetoresistance of an all-electrical spin-injection-detection device. The use of an oxide as a tunnel barrier has a few advantages. In comparison with Schottky tunnel barriers, where a ferromagnetic metal is used in direct contact with the semiconductor, the use of an oxide improves the chemical stability of the ferromagnet/semiconductor interface, which is often an issue in metal/semiconductor contacts. Furthermore, the resistance of the tunnel barrier should be easily adaptable, which can be realized by changing the thickness of the tunnel barrier, or using a different oxide with a different barrier height.

The band-structure of the AlO_x -based spin-LED under bias is shown in Fig. 4.1. Before the start of this research project, a spin-LED based on this principle had been developed in Imec.[2] The injector was a sputtered CoFe/AlO_x stack on an epitaxially grown $\text{AlGaAs}/\text{GaAs}$ spin-LED. The spin-LED showed a clear spin injection signal at 80 K, exhibiting a steady-state-spin polarization of 8%. At higher temperatures, however, the

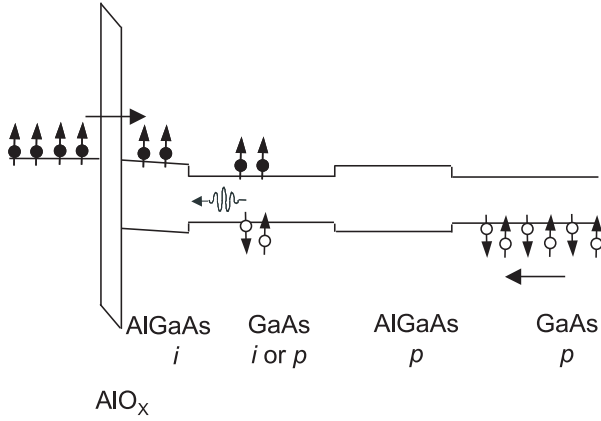


Figure 4.1: The schematic band profile of the metal-insulator-semiconductor-based spin-LED under bias.

enhanced spin relaxation reduces the steady state spin polarization, which complicates the determination of the injected spin polarization. Furthermore, at higher temperatures the optical efficiency dropped, again complicating a reliable measure of the spin polarization. In this specific structure the current density needed to obtain sufficient light emission was very high ($\sim 2000\text{A}/\text{cm}^{-2}$) at 80 K. The high current density needed for light emission is caused by the hole current, that freely flows from the substrate to the top contact at flatband conditions.

The choice for the CoFe/ AlO_x -stack was straightforward, based on the know-how in our research group on the fabrication of AlO_x in magnetic tunnel junctions for MRAM-purposes.

In order to improve the value of injected spin polarization and extend the results to room temperature, the fabrication of the AlO_x tunnel barrier was optimized and adapted to the properties of the substrate, which is an MBE-grown AlGaAs/GaAs heterostructure, in contrast to the sputtered magnetic metal electrode in the case of the magnetic tunnel junctions. In order to facilitate a full oxidation and to prevent the formation of pinholes, a two-step oxidation process has been developed for the use in magnetic tunnel junctions and has been transferred to the spin-LEDs.

Thickness	Material	Doping	Purpose
5 nm	Cu	—	Ferromagnetic metal
8 nm	NiFe	—	
2 nm	CoFe	—	
1.3 nm	AlO _x	—	Tunnel barrier
15 nm	Al _{0.2} Ga _{0.8} As	<i>i</i>	Transport region
100 nm	GaAs	<i>i</i> or $p = 10^{18}\text{cm}^{-3}$	Active region
20 nm	Al _{0.3} Ga _{0.7} As	<i>i</i> or $p = 2 \times 10^{18}\text{cm}^{-3}$	
180 nm	Al _{0.3} Ga _{0.7} As	$p = 10^{18}\text{cm}^{-3}$	
1 μm	GaAs	$p = 10^{18}\text{cm}^{-3}$	
Substrate	GaAs	$p = 10^{18}\text{cm}^{-3}$	

Table 4.1: The MIS-spin-LED structure.

4.2 Experimental results

In this experiment the polarization of the injected electrons also was monitored through a polarization analysis of the emitted light in a surface emitting metal-insulator-semiconductor (MIS) spin-LED using the oblique Hanle effect.

The structure of the fabricated spin-LEDs is depicted in Table 4.1. The semiconductor heterostructure was grown by molecular beam epitaxy. Immediately after the growth and the unloading from the vacuum, the sample was transferred through air into the sputtering machine for the fabrication of the tunnel injector. The oxide layer was deposited in a two-step process, which facilitates a full oxidation of the Al, reduces the chance on pinholes[3] and enables the fabrication of a thicker barrier. First, a 1 nm thin Al layer is sputtered and naturally oxidized in a controlled oxygen atmosphere at 140 Torr. After sputtering and oxidation of the first Al layer, a second Al layer was sputtered and oxidized, followed by the deposition the ferromagnetic stack. All metals are dc magnetron sputtered. In comparison with ref[8], the thickness of the AlO_x-layer is increased to allow a better optical efficiency due to the relative change of the tunnel probabilities for electrons and holes. This enables functioning of the device at lower operating currents. Now, typical operation currents of 400 to 600 A/cm² at 80 K are needed for sufficient light emission. This allows us to perform optical measurements at higher temperatures and at lower current stress applied to the tunnel oxide. Moreover recent calculations[1] suggest that enlarging the spin-dependent interface resistance induced by the tunnel barrier should result in an increase of the spin injection efficiency. The transmission electron microscopy (TEM) image of the MIS-structure (Fig. 4.2) shows that

the tunnel-oxide is smooth, exhibits no pinholes and has atomically sharp interfaces with both the AlGaAs and the ferromagnet.

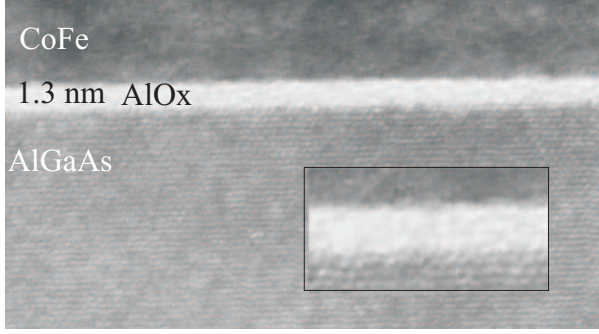


Figure 4.2: Transmission Electron Microscopy (TEM) picture of the CoFe-AlO_x-AlGaAs interface of the fabricated spin-LED. In the inset a magnification of the interface is shown.

Under forward bias spin polarized electrons are injected from the ferromagnetic top layer into the undoped 100 nm wide GaAs active region. There they radiatively recombine with holes supplied by the substrate. The AlGaAs barriers reduce the non-radiative surface recombination, confine the electrons and holes in the active region and thus further improve the optical efficiency. The measured spectrum of the electroluminescence showed that band-edge interband transitions provided the dominant contribution to the light emission. We can hence apply the selection rules to obtain reliable information on the spin polarization of the injected carriers.

4.2.1 The spin polarization of the injected carriers.

The results discussed in this section are measured on devices with an undoped active region. The analysis of the measured degree of circular polarization is performed in the oblique Hanle effect geometry. In our structure, the angle of the magnetization with the plane θ shows an approximately linear behavior as a function of the applied magnetic field and reaches about 10° at 0.6 T (A linear behavior was observed in Alternating Gradient Field Magnetometer (AGFM) and Magnetic Circular Dichroism (MCD) measurements in an oblique magnetic field). Extraordinary Hall Effect (EHE) measurements have shown that the out-of-plane magnetization of the thin film saturates at $B_\perp = 1.3$ T. A calculation using eq. 3.6 results in an angle of 10° at 0.6 T.

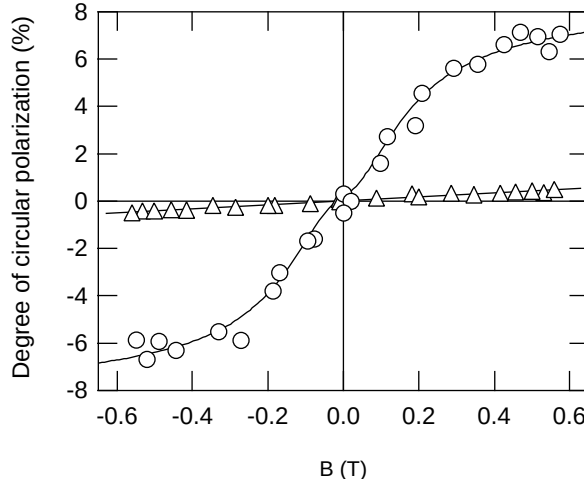


Figure 4.3: Degree of circular polarization as a function of the oblique magnetic field in the MIS spin-LED (circles) with an undoped active region at 80 K, together with the MCD-signal ($\circ$) for a device with an undoped active region (The electroluminescence polarization signal has been corrected for the MCD-effect) and the OHE (eq. (3.4)) fit (solid line) with following fit parameters: Spin polarization in the semiconductor at 80 K: $\Pi_{inj} \cdot T_S/\tau = 24 \pm 3\%$ and spin lifetime at 80 K: $T_s = 0.14 \pm 0.01 \text{ ns}$.

Fig. 4.3 ($\circ$) shows the degree of circular polarization of the emitted light in our devices as a function of the oblique magnetic field at 80 K. The curve is strongly non-linear, characteristic for the OHE effect. The contribution of the MCD appearing in the ferromagnetic film and caused by the out-of-plane component of the magnetization of the thin magnetic film is measured in a completely optical experiment and has been subtracted from the measured signal.

The fit using eq.(3.4) gives a spin polarization $\Pi_{inj} \cdot T_s/\tau = 24 \pm 3\%$ and a spin lifetime $T_s = (140 \pm 10) \text{ ps}$. The actual spin polarization of electrons that traversed the ferromagnetic metal / semiconductor interface $\Pi_{inj} = \tau/T_s \cdot 24\%$, is higher than 24% by the factor τ/T_s . The parameter T_s/τ describes the spin scattering of electrons during their lifetime on the bottom of the conduction band in the semiconductor. The T_s/τ ratio is not known for the active region of the device since the supporting optical injection-detection experiment is not possible (the PL is dominated by the highly doped GaAs substrate).

The room temperature results are presented in Fig. 4.4 after subtrac-

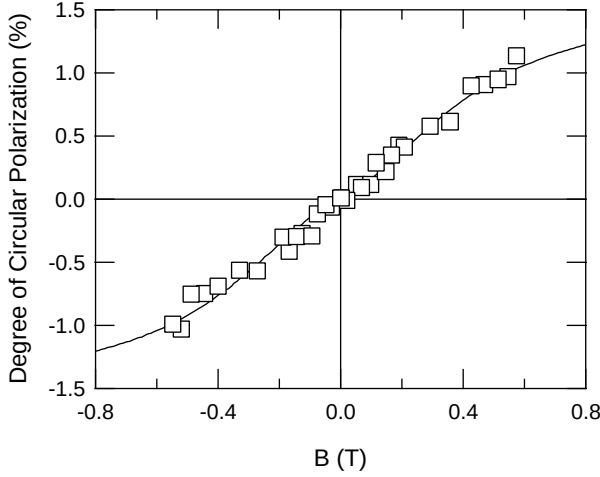


Figure 4.4: Degree of circular polarization as a function of the oblique magnetic field in the MIS spin-LED at room temperature for a device with an undoped active region (The signal has been corrected for the MCD-effect) and the OHE fit (following eq. (3.4)) (solid line) with following fit parameters: Spin polarization in the semiconductor at 300 K: $\Pi_{inj} \cdot T_S/\tau = 4.7 \pm 1\%$ and spin lifetime at room temperature: $T_s = 55 \pm 20$ ps.

tion of the MCD-contribution. The measured curve now looks linear, as expected, since at room temperature the enhanced spin scattering leads to a broadening of the Hanle curve. Within the small range of the magnetic field this does not allow observation of saturation of the measured circular polarization. The most conservative Hanle fit reveals a spin polarization of $\Pi_{inj} \cdot T_s/\tau = (4.7 \pm 1) \%$ and a spin lifetime T_s of (55 ± 20) ps.

The difference in $\Pi_{inj} \cdot T_s/\tau$ and T_s at 80 K and RT is not surprising and entirely reflects the spin dynamics within the GaAs spin detector. It is well known[4] that the electron-hole recombination time τ slightly increases with temperature and that the spin relaxation time τ_S rapidly decreases with temperature. This is clearly confirmed by the observation of a reduction of the spin-lifetime with increasing temperature T_S : $(T_S)_{80K}/(T_S)_{300K} \approx 2.54$. Hence, the decrease of the spin lifetime T_S , as well as the increase of the electron lifetime τ both lead to a smaller value of T_S/τ at room temperature. In this case, assuming $T_S/\tau \sim 1$ at 80 K, the lower bound of polarization of electrically injected electrons is $\Pi_{inj} = 12 \pm 3 \%$ at RT.

4.2.2 Bias dependence of the injected spin polarization.

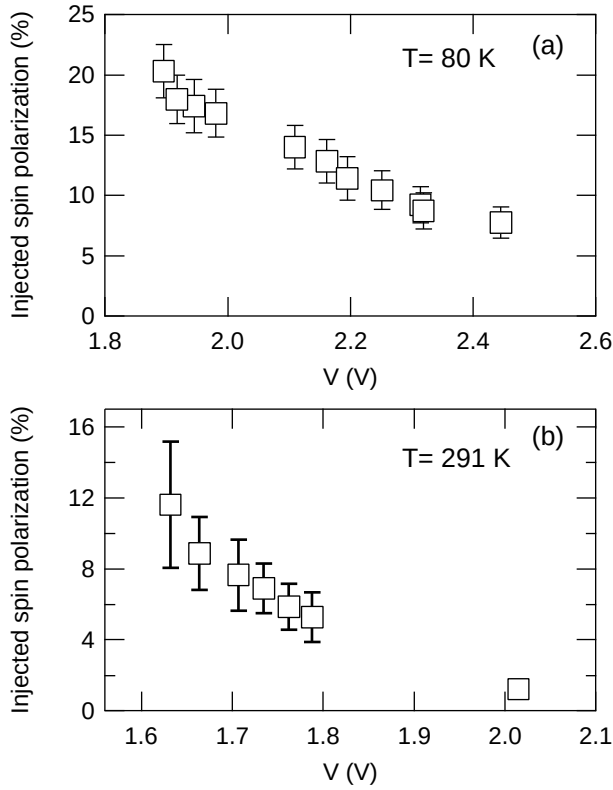


Figure 4.5: The injected spin polarization in the (undoped) GaAs active region as a function of the bias voltage at 80 K (a) and at 291 K (b).

In Fig. 4.5, the bias dependence of the measured spin polarization of a device with an undoped active region is shown, both at 80 K and at RT. It can be clearly seen that the spin polarization decreases for a higher applied bias voltage. The decrease of the injected spin-polarization with the applied bias is interesting, as it reminds of the bias dependence of the TMR in magnetic tunnel junctions.[6] This bias dependence is usually attributed to inelastic tunneling processes, involving the emission of magnons or tunneling through defect states. The former effect is unlikely in our experiment, since the positively biased electrode (the semiconductor in our case) is not ferromagnetic and the oxide does not (should not) contain magnetic impu-

rities. At this stage we cannot rule out the second effect, but we can argue that it is very unlikely to occur, giving the very thin oxide used in the experiment. From the semiconductor point of view, the bias dependence of the polarization can have another explanation. Kioseoglou et al showed in an adapted spin-LED structure with a bulk undoped active region, similar to the one employed in our experiment, that at low bias a two-dimensional hole gas (2DHG) is created near the GaAs/AlGaAs-interface [5]. The confinement in this 2DHG lifts the degeneracy of the valence band and changes the selection rules for interband transitions. They indeed observed a decrease of the light polarization with the applied bias (the polarization at high bias was exactly half of the low-bias polarization, directly reflecting the changing selection rules) and a shift of the electroluminescence spectrum. The effect rapidly decreased with increasing temperature, to disappear near 13 K, as a result of the thermal spreading of the holes in the entire active region.

A conclusive explanation for the observed decrease of the steady-state spin polarization is provided by the DP-mechanism for spin relaxation of conduction electrons. As mentioned in Chapter 2, the efficiency of the DP-spin relaxation mechanism is strongly enhanced at higher energy in the conduction band, especially for undoped GaAs. As the applied voltage creates hot electrons, the DP-mechanism can cause the injected spin polarization to vanish before reaching the bottom of the conduction band.

In order to examine the origin of this peculiar bias dependence of the steady-state spin-polarization self-consistent simulations of the total spin-LED structure have been performed using Medici, as to determine where the voltage drop takes place, while biasing the spin-LED. Special care has been taken to include the Fermi level pinning at the AlGaAs/ AlO_x -interface in the simulations, as this has a great impact on the simulation results. The self-consistent simulations of the band profiles in Fig. 4.6 clearly indicate that the bands in the active GaAs region in the spin-LED are flat in the entire relevant range of the applied bias voltage, excluding the confinement effects reported by Kioseoglou. Furthermore, the measurement temperature is well above 13 K (80 K and RT).

In order to examine the possible effects of defect states in the AlO_x tunnel barrier and the spin-relaxation by hot electrons, a reference sample has been fabricated, with the same structure, except for the doping level in the active region, which has been chosen to be $2 \times 10^{18} \text{cm}^{-3}$. [7] The results, as shown in Fig. 4.7 indicate that using a heavily p-type doped active region essentially removes the bias dependence of the injected spin polarization. Since the oxide barrier has been fabricated by an identical procedure as in the case of the undoped active region, it is highly unlikely that defect states play an important role in the bias dependence of the injected spin polarization.

The difference of the bias dependence for the sample with the undoped

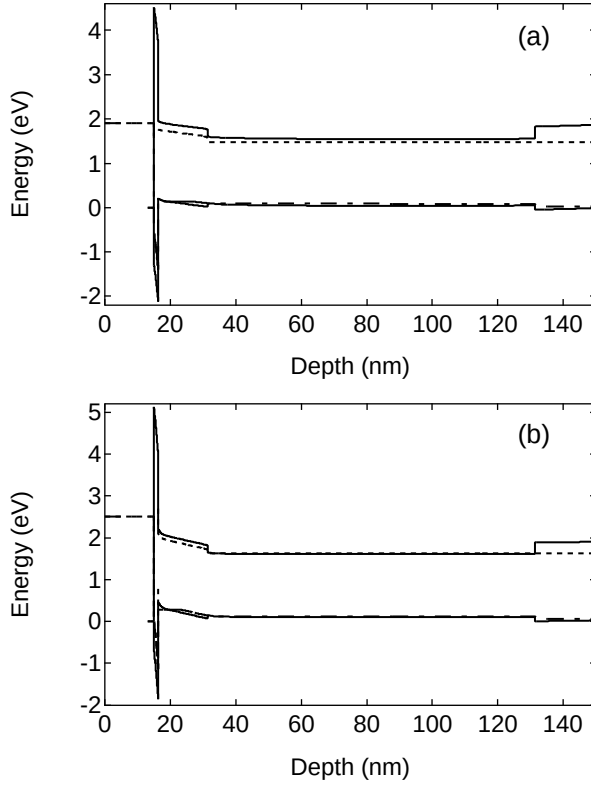


Figure 4.6: The band profile of the spin-LED with an undoped active region at 80 K under a bias of 1.9 V (a) and at 2.5 V (b). The dotted and the dashed-dotted lines represent the quasi-Fermi levels for electrons and holes, respectively.

active region and the highly doped active region can be explained in the framework of the D'yakonov-Perel spin relaxation mechanism whose efficiency strongly increases with energy.[8; 9] Due to the lack of inversion symmetry in GaAs, the conduction band is spin-split. The splitting depends on the energy ($\sim (E - E_C)^{3/2}$). This splitting acts as an effective magnetic field on the spins, which depends on their quasi-momentum and will cause the spins to precess. The relevant time scale of this precession is the relaxation time to the bottom of the conduction band. In undoped GaAs, this is the LO-phonon time τ_{LO} . This time is long enough to cause a randomization of the spins before they reach the bottom of the conduc-

tion band and leads to a reduction of the spin polarization at high applied bias. For highly doped GaAs, the characteristic timescale of ionized impurity scattering τ_{ip} is smaller than τ_{LO} . Before the electron thermalizes it undergoes several momentum scattering events. When this scattering time is small enough, a dynamical narrowing of the precession angle occurs and the spin is conserved, resulting in a nearly constant spin polarization as a function of the applied bias. This is schematically shown in Fig. 4.8

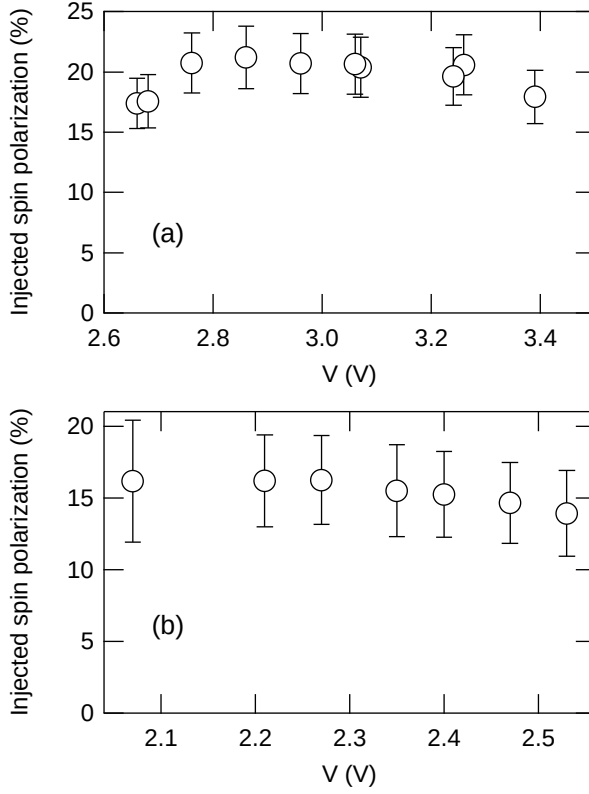


Figure 4.7: The injected spin polarization in the GaAs active region with a heavily p-type doped active region as a function of the bias voltage at 80 K (a) and at 291 K (b).

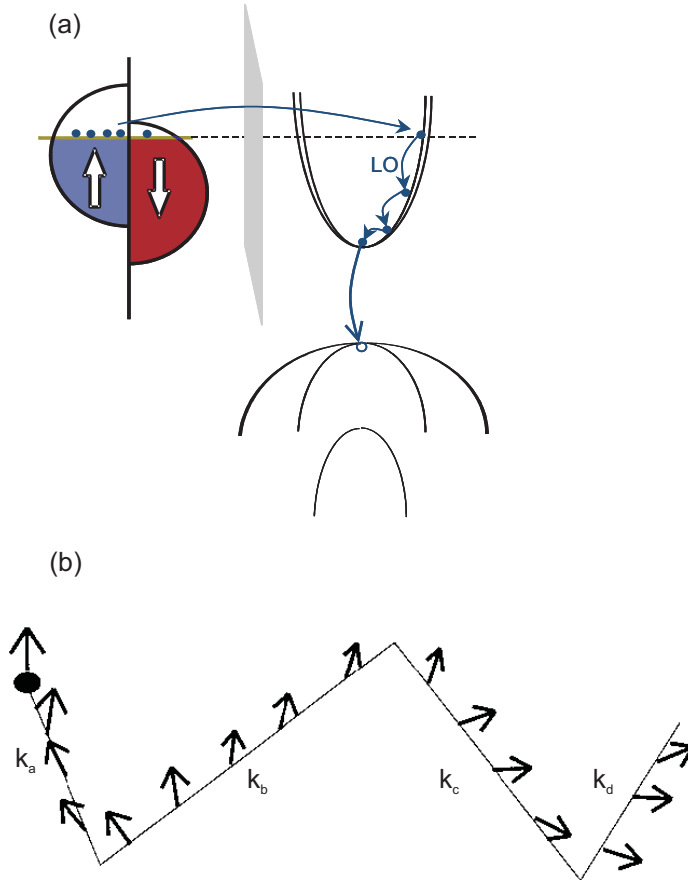


Figure 4.8: (a) At high bias electrons are injected high in the GaAs conduction band, where they thermalize by emitting LO phonons. (b) A schematic picture of the DP-spin relaxation mechanism as adopted from ref.[10]: the momentum-dependent effective magnetic field causes the electron spins to acquire a precession angle during their stay at one point in k-space. When the electrons scatter fast enough between different k-states, dynamical narrowing of the precession cone occurs and spin relaxation is suppressed.

4.2.3 Bias dependence of the width of the Hanle curve.

The width of the Hanle curve $\Delta B = (\frac{g^* \mu_B}{\hbar} T_S)^{-1}$ is inversely proportional to both the effective g-factor and the spin-lifetime in the active region. In Fig. 4.9 is shown that the width of the Hanle curves for the sample with

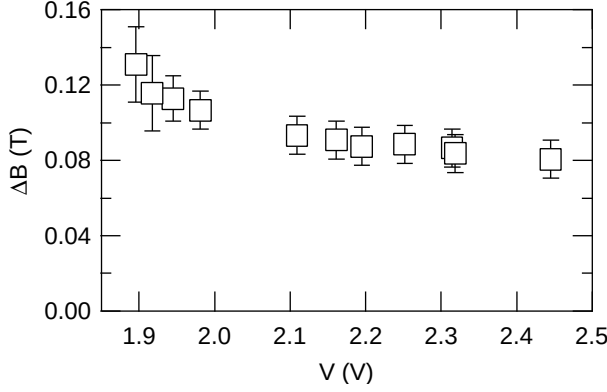


Figure 4.9: The width of the Hanle curve as function of the applied bias voltage at 80 K for the sample with the undoped active region.

the undoped active region decreases monotonically with bias at 80 K. For the sample with the doped active region an identical dependence has been found at 80 K. At room temperature, no dependence of the width on the applied bias has been observed for both samples. This seems to suggest that at low temperature the spin lifetime increases with the applied bias. However, first-order effects seem to call for a decrease of the spin-lifetime with the applied bias instead of an increase: the heating of the carriers and the lattice would rather result in a decreased lifetime. One could also imagine that at low bias, before flat-band conditions the built-in electric field would separate the electrons and the holes. This, however, also would lead to a decrease of the spin-lifetime with bias.

Another potential explanation would be a change in the effective g-factor since the width is also inversely proportional to the effective g-factor. The triangular distortion of the active region at low bias caused by the built-in electric field can cause the electron wave function to penetrate in the AlGaAs barrier. Since the effective g-factor in GaAs is -0.44 and in the AlGaAs barrier it is 0.23,[11] this would cause a lower effective g-factor at low bias. The result of the self-consistent simulations of the band profiles, as shown in Fig. 4.6 indicate however that the active region is flat in the entire relevant voltage range. This excludes any effect of the AlGaAs region on the g-factor.

A third possibility would be that the thermalization is rather slow and the spins undergo some precession before they reach the bottom of the conduction band. However, a fully optical pumping experiment, using a higher excitation energy (1.96 eV excitation from a HeNe-laser) for the

injected spins, doesn't show a smaller width of the Hanle curve.

This leaves only dynamical nuclear polarization (DNP) as a good explanation for the observed behaviour. At higher biases, more electrons are injected and the injected electron spins can dynamically polarize nuclear spins due to the hyperfine interaction. The polarized nuclei induce, again through the hyperfine interaction, an effective magnetic field that accelerates spin precession. At higher biases more nuclear spins are polarized and the effective field is higher, resulting in a narrowing of the Hanle curves. The interaction between electron and nuclear spins will be studied in more detail in Chapter 8.

4.3 Conclusion

In conclusion, we have shown large spin injection efficiencies for spin injection in a MIS-LED structure at low and at room temperature by optimizing the thickness and the structural parameters of the tunnel barrier. The inherent properties of the ferromagnet- AlO_x -semiconductor interface (thermodynamical stability, no lattice mismatch problems), the robustness of the fabrication process and the use of the well-established magnetic tunnel junction (MTJ) technology demonstrate the potential of MIS injectors for other spintronic devices. Following our work, the results presented in this chapter have recently been confirmed by van 't Erve *et al*[12] who used a similar structure and obtained a maximum steady state spin polarization of 40% at 4.5 K. Furthermore, using MgO as a crystalline tunnel barrier between the AlGaAs/GaAs spin-LED and the CoFe injector, Jiang *et al* obtained a steady state spin polarization of 52% at 100 K and 32% at room temperature, demonstrating the prospects of oxide-based spin injectors[13]. The high obtained spin polarization in the latter report originates from the different damping of the wavefunctions of spin-up and spin-down electrons in the MgO-bandgap.

Furthermore, we have examined the bias dependence of the injected spin polarization and by comparing samples with a different doping level and using self-consistent simulations of the device under bias we were able to explain the bias dependence in terms of the DP-spin relaxation mechanism. The bias dependence of the width of the Hanle curve indicates a nuclear spin polarization in the GaAs active region.

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Chapter 5

Spin injection in a Schottky-based-spin LED

5.1 Introduction

The native Schottky barrier that is formed at a metal-semiconductor interface can be engineered in order to supply a spin-dependent tunnel resistance, necessary for efficient spin injection. When the depletion region of the Schottky diode is sufficiently narrow, tunneling between the ferromagnetic metal and the semiconductor becomes the dominant transport process. Since the ferromagnetic metal and the semiconductor are in direct contact, a good chemical stability of the interface is a prerequisite for reliable device operation. As mentioned in Chapter 3, some groups have demonstrated electrical spin injection using an epitaxially grown Fe metal/Schottky barrier combination. The growth temperature of the Fe is of crucial importance, since Fe-layers grown at high temperature on GaAs can show intermixing with the GaAs substrate and magnetic dead layers are formed. Therefore, the Fe layers are typically grown at room temperature, resulting in a better interfacial quality. The current record value of spin injection from Fe into GaAs is held by Hanbicki *et al* (30% at 4.5 K).[1] In this case the semiconductor was heavily doped near the Fe/GaAs interface, i.e. the doping level was ramped up steeply from $1 \times 10^{16} \text{cm}^{-3}$ to $1.5 \times 10^{19} \text{cm}^{-3}$, creating a narrow tunnel barrier. A similar result can also be obtained by introducing a delta-doping layer of Si, a few nanometers below the metal/semiconductor interface. Strand *et al* employed this technique, and successfully demonstrated spin injection, although with a somewhat smaller value.[2]

In this Chapter, we will investigate electrical spin injection from epitaxially grown MnSb into an AlGaAs/GaAs spin-LED, focussing on the

bias dependence of the injected spin polarization. Furthermore, the MnSb-injector exhibits interesting magnetic properties such as square hysteresis loops, with 100% remanence and a high coercivity, allowing Hanle curves with characteristics from figure 3.4(d). The studied devices exhibit also interesting effects dealing with the hyperfine interaction between the electronic and nuclear spins. This will be explored in more detail in Chapter 8

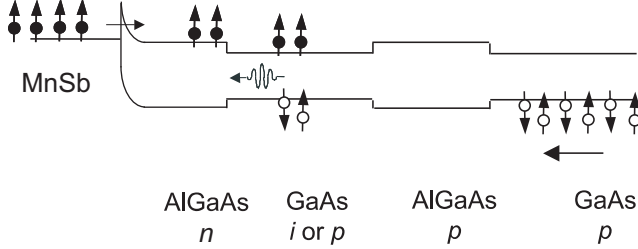


Figure 5.1: The schematic band diagram of the spin-LED with a MnSb/Schottky injector under bias. The doping profile in the AlGaAs top layer is designed to create a narrow depletion region, allowing electrons from the MnSb Fermi level to tunnel into the AlGaAs conduction band, where they are swept to the GaAs conduction band by the electric field. In the GaAs active region they recombine with holes supplied by the p-type substrate.

5.2 Experimental

Spin injection from epitaxial MnSb layers into GaAs through a Schottky-barrier has been studied. Since the surface Fermi level pins around the middle of the (Al,Ga)As band gap, we need to heavily dope the semiconductor region in the vicinity of the interface to obtain a tunneling Schottky barrier. The band structure and the doping profile of the LED were designed such that under bias the spin-polarized electrons tunnel through the highly n-type doped (Al,Ga)As interfacial region travel through a lightly n-type doped (Al,Ga)As region to a GaAs active region where they thermalize and recombine with holes supplied from the p-type substrate. The structure is depicted in table 5.1 and the band profile under bias is shown in Fig. 5.1.

The active region in this LED is 100 nm wide undoped GaAs. This is the same as in sample A of the AlO_x -injectors. MnSb is a ferromagnetic material that is particularly suited to study the effects of the magnetization on the Hanle characteristics. It has the hexagonal NiAs crystal structure resulting in a strong magnetocrystalline anisotropy with the c-axis as hard

Thickness	Material	Doping	Purpose
5 nm	GaAs	–	Cap layer
10 nm	MnSb	–	
8 nm	Al _{0.1} Ga _{0.9} As	$n = 1.5 \cdot 10^{19} \text{cm}^{-3}$	Tunnel barrier
5 nm	Al _{0.1} Ga _{0.9} As	$n = 7.5 \cdot 10^{18} \text{cm}^{-3}$	Tunnel barrier
10 nm	Al _{0.1} Ga _{0.9} As	$n = 3 \cdot 10^{18} \text{cm}^{-3}$	
20 nm	Al _{0.1} Ga _{0.9} As	$n = 3 \cdot 10^{17} \text{cm}^{-3}$	Transport region
20 nm	Al _{0.1} Ga _{0.9} As	$n = 5 \cdot 10^{16} \text{cm}^{-3}$	Transport region
100 nm	GaAs	i	Active region
20 nm	Al _{0.3} Ga _{0.7} As	i	
180 nm	Al _{0.3} Ga _{0.7} As	$p = 10^{18} \text{cm}^{-3}$	
1 μm	GaAs	$p = 10^{18} \text{cm}^{-3}$	
Substrate	GaAs	$p = 10^{18} \text{cm}^{-3}$	

Table 5.1: The Schottky-barrier based spin-LED structure.

direction and the basal plane as easy plane. For a substrate temperature of 250 C MnSb grows on GaAs(001) in the $(\bar{1}\bar{1}01)$ orientation, with the projection of the c -axis aligned along the GaAs[110] direction.[3] The shape anisotropy requires the easy magnetization direction(s) to be in the plane of the film. The magnetocrystalline anisotropy of MnSb reduces the symmetry within this plane and results in an easy in-plane direction along $[\bar{1}\bar{1}0]$, and a hard direction orthogonal to this, along $[110]$. This is clearly shown by the in-plane magnetization loops measured with an alternating gradient field magnetometer (AGFM) parallel to the $[\bar{1}\bar{1}0]$ and the $[110]$ -direction (Fig. 5.2). The $[\bar{1}\bar{1}0]$ easy direction exhibits square loops with a relatively large coercive field, which allows us to visualize the effects of the magnetization on the Hanle curve. An out-of-plane AGFM measurement (not shown) showed a hard-axis behavior with the saturation magnetization of the film equal to $\mu_0 M_S = 0.8$ T.

The EL-spectrum of the spin-LED is shown in Fig. 5.3. The main contribution to the spectrum is again given by band-edge interband transitions. Near a wavelength of 835 nm a second peak appears in the electroluminescence spectrum. This could be caused by electrons recombining in the p-type substrate or by an impurity-related transition in the active region. Since the relative intensity of the 835 nm-feature is bias-independent, we can assume that the feature is impurity-related. The selection rules of such a transition may differ from the bulk transition rules, but generally lead to a lower estimate of the spin polarization, as reported by Jonker *et al.*[4] Hence, the degree of circular polarization of the electroluminescence, measured by integrating the entire spectrum is a lower estimate of the total

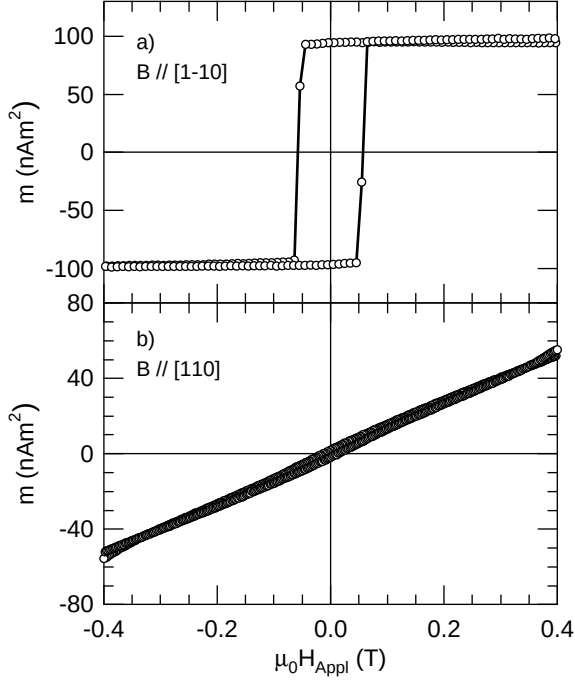


Figure 5.2: (a) The magnetization of the MnSb film as a function of the magnetic field in the $[1\bar{1}0]$ direction. (b) The magnetization of the MnSb film as a function of the magnetic field in the $[110]$ direction

degree of circular polarization. However, since the band-edge transitions dominate the spectrum, the circular polarization is not expected to differ much from the actual value.

The OHE-technique has been employed to analyze the circular polarization of the light. The measured circular polarization is shown for different values of the applied bias voltage as a function of the applied magnetic field in Fig. 5.4(a). The in-plane component of the magnetic field in this measurement was oriented along the easy axis of the magnetization. The threshold for luminescence was around 1.47 V. At this low bias however, no signal was detected that can originate from spin injection. The measured circular polarization shows a weak linear dependence with the applied magnetic field and exhibits no hysteresis in the low field range. The linear slope

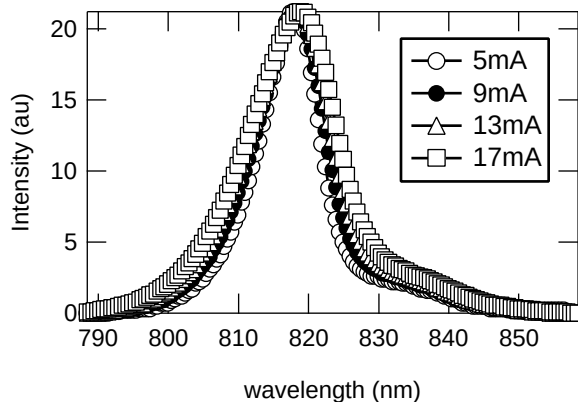


Figure 5.3: The spectrum of the electroluminescence of a Schottky-spin-LED for different values of the applied current.

originates from MCD (magnetic circular dichroism) in the MnSb film. This low-bias measurement immediately sets a lower limit to any parasitic circular polarization that can be acquired. This has been subtracted from the measured circular polarization at other bias voltages. In Fig. 5.4(b) the corrected Hanle curve for a bias of 2.33 V is shown. In this graph the low-bias background polarization has been subtracted from the raw data. The evolution of the degree of circular polarization as a function of the magnetic field corresponds qualitatively with Fig. 3.4. When the magnetic field is swept from positive to negative values, one sees that the degree of circular polarization decreases at low fields, increases again when the field reverses its sign and finally switches to negative values when the magnetization of the MnSb-film reverses.

The steady-state spin polarization in the GaAs active region has been obtained for the different bias voltages by fitting the curves with eq. 3.4 and is shown as function of the bias voltage in Fig. 5.5. It is clear that the spin polarization exhibits a very strong bias dependence. This bias dependence is quite different from the one of the AlO_x -injector with the same active region. In order to examine the bias dependence of the charge distribution and the band bending in the spin-LED, self-consistent simulations have been performed of the entire spin-LED structure. The obtained band profiles are shown in Fig. 5.6. It can be observed that at low bias the (Al,Ga)As spin-transport region is not fully depleted and there is a retarding electric field for electrons injected from the ferromagnetic electrode. This will cause an increase of the transit time for injected electrons. Furthermore, the high

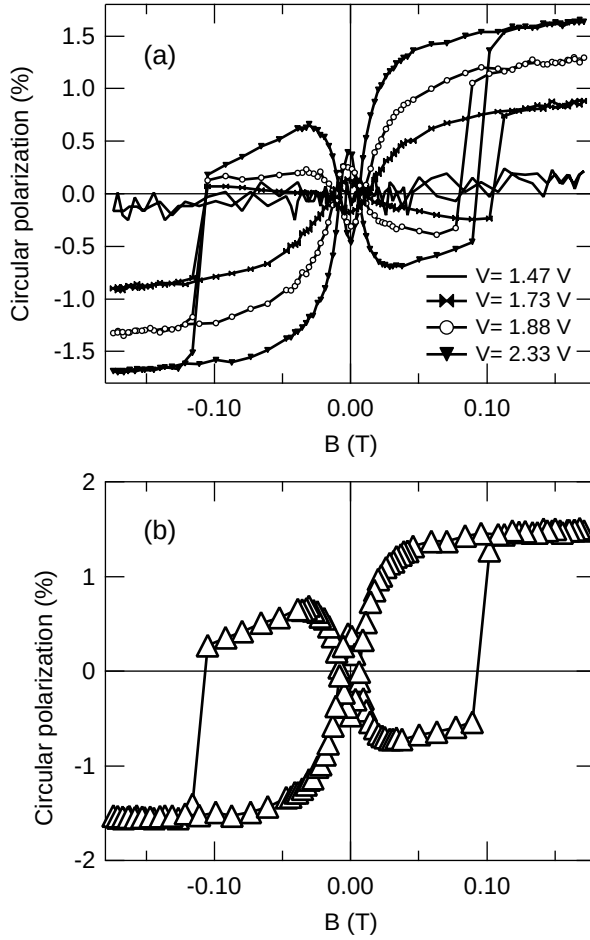


Figure 5.4: (a) The raw Hanle curves for different values of the applied bias. (b) the Hanle curve at 2.33 V after subtraction of the low-bias background polarization.

electron concentration in the (Al,Ga)As transport region causes the spin-relaxation rate to increase, as the DP-spin relaxation mechanism dominates. The increased transit time, combined with efficient spin-relaxation quenches the spin-polarization of the injected carriers, before they reach the active GaAs region. Furthermore, it can be seen in Fig. 5.6(a) that there is an

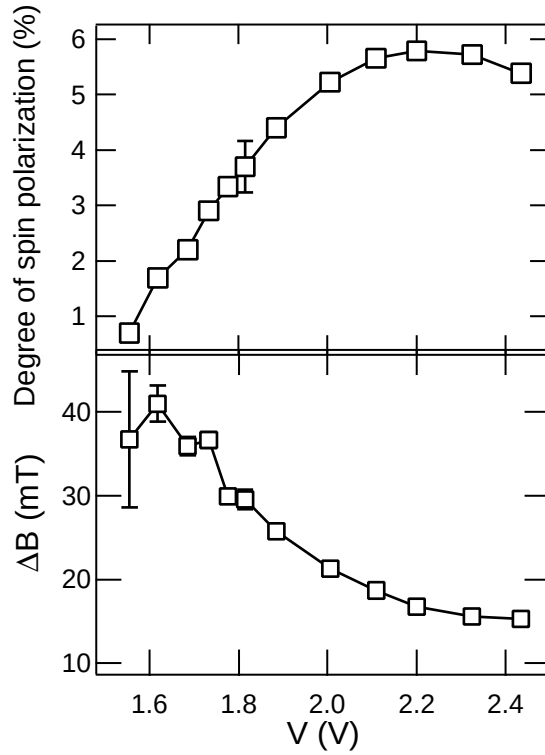


Figure 5.5: The bias dependence of the steady-state spin polarization in the GaAs active region (a) and the width of the Hanle curves (b) at 80 K.

electric field present in the active region, separating electrons and holes and hence increasing the electron-hole recombination time, resulting in a reduced factor T_S/τ . With increasing bias the transit time is drastically decreased due to depletion of the (Al,Ga)As region and a reduction of the retarding electric field. Furthermore, the GaAs active region is flat, allowing efficient recombination between electrons and holes. As a consequence a higher polarization has been measured in the GaAs active region. In Fig. 5.5(a) one can also observe that above 2.2 V the signal decreases again. We can attribute this to an enhanced spin relaxation due to the D'yakanov-Perel (DP)-mechanism, which is more effective at higher energy.

Fig. 5.5(b) shows the dependence of the width of the Hanle curves on the applied bias. The measured Hanle curves at the highest bias are 10 times narrower than the Hanle curves in the case of sample A although in

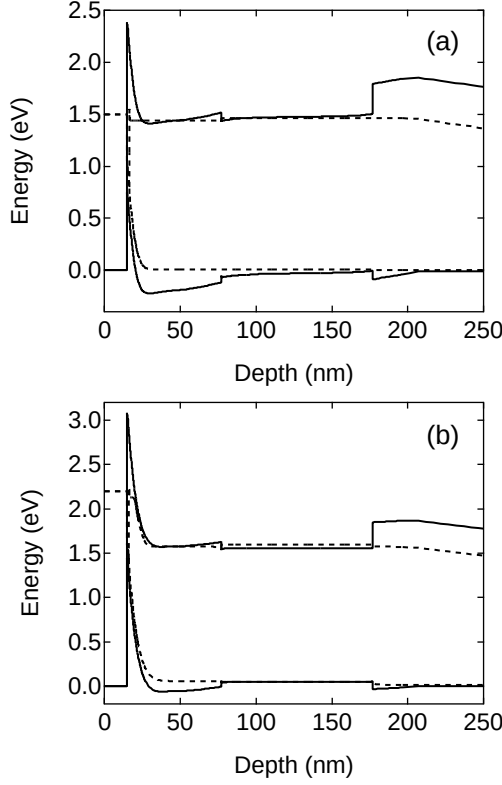


Figure 5.6: The band profiles of the spin-LED's under a bias of 1.5 V (a) and 2.2 V (b) at 80 K.

both cases the active region is undoped. Furthermore, as in the case of the AlO_X -spin LEDs, the width depends on the applied bias. The enhanced width could be explained by spin precession in the AlGaAs spin-transport region. However, applying a higher bias should reduce the time the electrons spend in this region, while in this case at higher biases the width decreases. Furthermore, the electron-hole separating electric field, which is present in the GaAs active region at low bias, should result in a decreased width at low bias. Increasing the bias and creating a flatband condition should result in an increase of the width. Like in the case of the AlO_X -spin-LEDs, dynamical nuclear polarization is proposed as a possible cause of this phenomenon. The injected electron density is substantially higher than in the case of the AlO_X -spin LED's where the measured light intensity

is significantly lower. The higher electron concentration creates a larger nuclear polarization, which in turn create a higher effective magnetic field. Another indication of nuclear polarization is shown in Fig. 5.4(b). When lowering the field from 0.2 T, the circular polarization goes down, has a minimum when the field goes through zero, increases, but decreases again before the magnetization reverses. Normally, the curve should be symmetric round zero (as in Fig. 3.4(d)), which is not the case here, even when the parasitic MCD-signal has been subtracted from the curve and the tilting of the magnetization out of plane has been taken into account. When we assume nuclear polarization, the Hanle curve should be shifted with the nuclear field B_N and around zero field a sharp dip should occur due to quenching of the nuclear polarization at zero field. Chapter 8 will deal with this phenomenon more extensively.

5.3 Conclusion

In this chapter we have demonstrated electrical spin injection in an Al-GaAs/GaAs spin-LED using a tunneling process from a ferromagnetic MnSb top contact through the tailored Schottky barrier at the MnSb-AlGaAs interface. We have measured the bias dependence of the steady-state spin polarization and have explained it using self-consistent simulations of the entire device under bias. Furthermore the bias dependence of the width of the Hanle curve gives indications of a nuclear spin polarization in the GaAs active region

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Chapter 6

Electrical spin injection in (Ga,Mn)As-based spin-LEDs

6.1 Introduction

Spin injection is one of the target applications of ferromagnetic semiconductors, which can serve as a natural supply of highly spin-polarized carriers. In particular, the most intensively studied ferromagnetic semiconductor (Ga,Mn)As can be grown epitaxially on GaAs and the carrier-mediated ferromagnetism in this material provides an elegant way to control the ferromagnetic properties by tuning the hole concentration.[1] A disadvantage of III-V based ferromagnetic semiconductors for spin-transport devices is their p-type character. The strong spin-orbit interaction in the valence band of the III-V semiconductors results in a very low hole spin-lifetime, in the order of the momentum scattering time (sub-picoseconds). Nevertheless, (Ga,Mn)As has been integrated as a hole-spin injector in spin-LEDs in previous experiments.[2; 3] An injected hole spin polarization up to 7% has been observed for spin-LEDs with the active region positioned within 20 nm of the injector layer. Increasing the spacer width between the injector and the active region resulted in a rapid quenching of the measured spin polarization. Conduction electrons are preferred as spin-carriers. Ultimately an n-type ferromagnetic semiconductor could be used to realize electron spin injection. Initial experiments have been performed at the Naval Research Lab in Washington D.C. on the epitaxial growth of the n-type ferromagnetic semiconductor CdCr_2Se_4 on GaAs and the incorporation of

this material in a spin-LED.[4] An injected spin polarization of 6% has been achieved using this material combination. The (III,Mn)V-semiconductors can, however also be employed as electron injectors. Recent experiments in Tohoku University, Japan and the UC in Santa Barbara, USA have shown that interband (Zener) tunneling from valence band electrons of (Ga,Mn)As to the conduction band of GaAs is a way to accomplish this.[5; 6] In both experiments the spin polarization was measured through the circular polarization of the light emitted from an InGaAs quantum well and injected spin polarizations of respectively 6.5% and 1% were found.

6.2 Experimental results.

We investigate interband tunneling from (Ga,Mn)As into an (Al,Ga)As light emitting diode (LED) as a route for efficient spin injection. We achieved a steady-state spin polarization of 60% in the GaAs active region at 4.5 K, which corresponds to an injected spin polarization of 80% when corrected for spin scattering in GaAs. This is the highest reported value of spin injection from a ferromagnetic material. The injected electron spin polarization decreased with temperature to vanish at the Curie temperature of the (Ga,Mn)As layer, 120 K. In addition the injected spin polarization is very sensitive to the applied bias - in contrast to former studies. We correlate this to the electrical properties of the spin-LED. This investigation deviates from the former ones in three ways. Firstly we use a thin, low-temperature annealed (Ga,Mn)As layer as injector. It has been shown that low-temperature annealing of thin (Ga,Mn)As films results in greatly improved material quality by the removal of Mn_I interstitial atoms, which act as double donors, from the films. This results in much reduced compensation, high hole concentrations and increased T_C 's up to 173 K.[7] This, together with the moderate n-type interfacial doping leads to a smaller depletion and hence a smaller demagnetization of the (Ga,Mn)As interface. Secondly the active region where the injected electrons radiatively recombine with holes and thus reveal their spin orientation is a 100 nm wide GaAs active region. We didn't use (In,Ga)As as spin detector because of its low spin lifetime and to avoid strain related parasitic sources of circular polarization. Finally the Oblique Hanle Effect (OHE) technique was used to measure the spin polarization. The spin-LED's were grown by molecular beam epitaxy and had the structure, as depicted in table 6.1.

After the growth of the LED the sample was As-capped and transferred to a second MBE-system used for the growth of magnetic materials. After removal of the As-cap, 20 nm $Ga_{0.92}Mn_{0.08}As$ was grown. From the temperature dependence of the resistivity of annealed (Ga,Mn)As-layers grown using the same growth conditions on semi-insulating substrates, as depicted

Thickness	Material	Doping	Purpose
20 nm	Ga _{0.92} Mn _{0.08} As	$p = 3.5 \times 10^{20} \text{ cm}^{-3}$	FM semiconductor
9 nm	GaAs	$n = 9 \times 10^{18} \text{ cm}^{-3}$	Tunnel barrier
90 nm	Al _x Ga _{1-x} As	$n = 1 \times 10^{17} \text{ cm}^{-3}$	Transport region
100 nm	GaAs	$p = 2 \times 10^{18} \text{ cm}^{-3}$	Active region
200 nm	Al _{0.3} Ga _{0.7} As	$p = 2 \times 10^{18} \text{ cm}^{-3}$	
1 μm	GaAs	$p = 1 \times 10^{18} \text{ cm}^{-3}$	
Substrate	GaAs	$p = 1 \times 10^{18} \text{ cm}^{-3}$	

Table 6.1: The (Ga,Mn)As-based spin-LED structure.

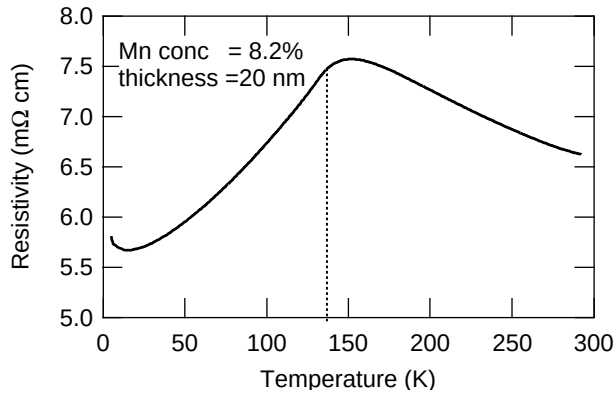


Figure 6.1: The resistivity of a (Ga,Mn)As-layer with a thickness of 20 nm, grown on a semi-insulating GaAs wafer as function of temperature. The sample has been annealed in a N_2 -atmosphere at 260°C for 1 hour. The peak of the resistivity-curve appears just above the Curie temperature of the (Ga,Mn)As film.

in Fig. 6.1 could be deduced that the T_C equals about 140 K after annealing at 260°C for 1 hour. The (Ga,Mn)As-layers in the spin-LED-structures exhibit a T_C of 50 K before and 120 K after annealing. After the growth the wafers have been processed into surface-emitting LED-devices.

When we apply a negative voltage on the (Ga,Mn)As electrode, the Zener diode is reverse biased while the LED-pn-junction is forward biased. The band structure under bias is shown schematically in Fig. 6.2. From about 1.6 V spin polarized electrons are injected from the valence band of (Ga,Mn)As into the conduction band of the AlGaAs spin-transport layer. The applied electric field drives them into the GaAs active region where

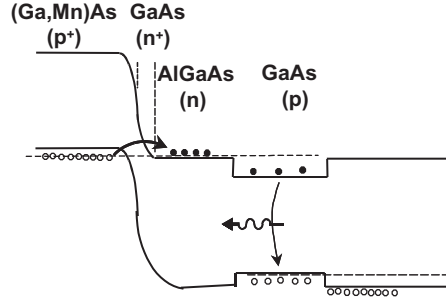


Figure 6.2: The band profile of the (Ga,Mn)As-spinLED under bias. The spin-polarized valence electrons in (Ga,Mn)As tunnel to the (Al,Ga)As conduction band, are swept to the GaAs active region, where they radiatively recombine with holes.

they thermalize and recombine with holes supplied from the p-type substrate. The spectra of both the photoluminescence and the electroluminescence show the signature of band-to-band recombination in the active region (Fig. 6.3). The semiconductor structure was designed and optimized using Medici, to avoid that injected electrons would spend too much time in a highly doped area where spin relaxation is very efficient.

The analysis of the circular polarization of the emitted light is performed using the OHE-technique. The magnetic field, needed to saturate the out-of-plane magnetization of (Ga,Mn)As, is small compared to most ferromagnetic metals. Therefore, the angle of the magnetization with the plane, induced by the oblique magnetic field, can be quite large. The in-plane anisotropy caused by the compressive strain in the (Ga,Mn)As layer and the shape anisotropy is characterized by the out-of-plane field H_{SAT} needed for saturating the magnetization of the thin film. This has been determined from SQUID-measurements in a perpendicular magnetic field showing a hard-axis behaviour with $\mu_0 H_{SAT} = 0.35$ T at 5 K, as indicated in Fig. 6.4. According to eq 3.6 at an oblique field of 0.54 T the angle of the magnetization with the plane is about 27° .

In order to quantify the electrical spin-injection experiments, some calibration experiments have to be performed. Since the emitted light travels through the (Ga,Mn)As thin film, we have to take into account the magnetic circular dichroism (MCD) effect, which can be strong in (Ga,Mn)As.[8] By illuminating the sample with unpolarized 1.96 eV laser light we generate unpolarized electron-hole pairs in the active region. The degree of circular polarization of the photoluminescence gives the parasitic MCD signal,

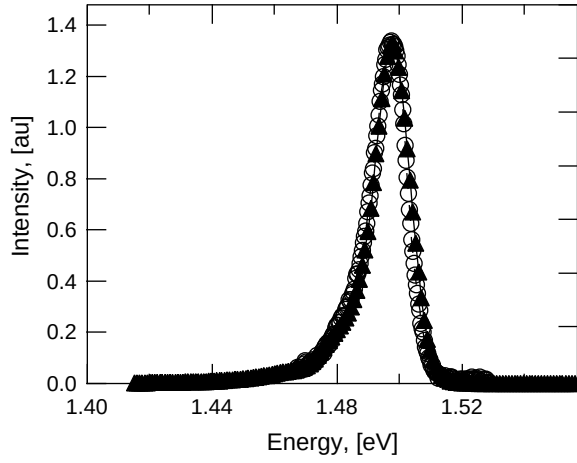


Figure 6.3: The spectrum of both the electroluminescence ($\circ$) and the photoluminescence ($\blacktriangle$) of the (Ga,Mn)As spin-LED.

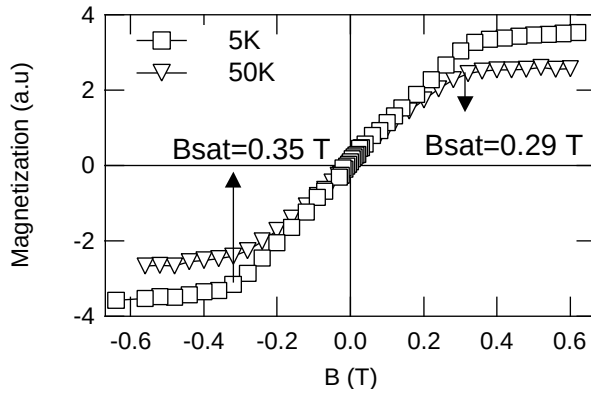


Figure 6.4: The out-of-plane magnetization of the (Ga,Mn)As thin film as function of the external magnetic field.

which is subtracted from the electrical spin injection measurements. The spin lifetime T_S and the factor T_S/τ , which is determined by spin scattering on the bottom of the conduction band in the GaAs active region, have also been obtained independently on the same sample in an optical pumping

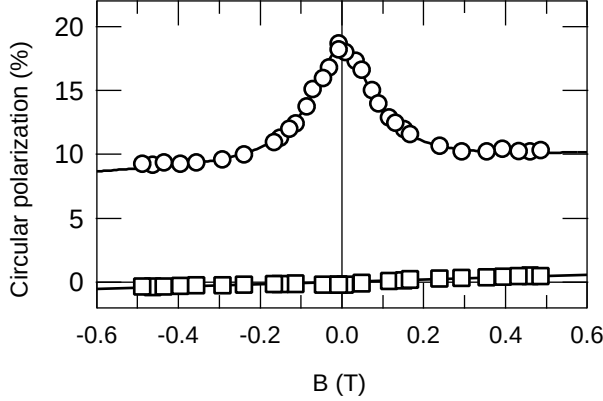


Figure 6.5: The circular polarization of the photoluminescence of the (Ga,Mn)As spinLED as function of the external magnetic field in the OHE-geometry for excitation with 1.58 eV circularly polarized light ($\circ$) and excitation with 1.96 eV unpolarized light ($\square$) at 4.6 K. The Hanle depolarization curve has been fit using the OHE-theory, resulting in a spin-lifetime $T_S = 290 \pm 20 ps$ and $T_S/\tau = 0.74$.

experiment. In Fig. 6.5 the degree of circular polarization is shown as a function of the applied magnetic field at 4.6 K. The measured values are $T_S = 290 \pm 20 ps$ and $T_S/\tau = 0.74$. T_S/τ and T_S also have been measured at elevated temperatures in order to fully characterize the spin relaxation parameters of the spin-detecting GaAs-layer in the relevant temperature range. The temperature dependence of T_S/τ is depicted in Fig. 6.6.

With the above parameters independently measured, one can now perform the electrical spin injection experiment. Fig. 6.7 shows the degree of circular polarization of the electroluminescence as a function of the applied magnetic field at 4.6 K. Fitting these results to Eq. 3.4, using only the injected spin polarization as a fit parameter, yields $\Pi_{inj} = 82 \pm 10\%$, the steady state spin polarization in the active region is in this case $= 60 \pm 6\%$. Fig. 6.8 shows the temperature dependence of Π_{inj} and of the (in-plane) remanent magnetization. Both vanish at 120 K, but the shape of the curves is different.

We have also measured the degree of injected spin polarization in non-annealed devices. In Fig. 6.9(a) the Hanle curve is shown at 4.6 K and at low bias. The extracted injected spin polarization Π_{inj} was measured to

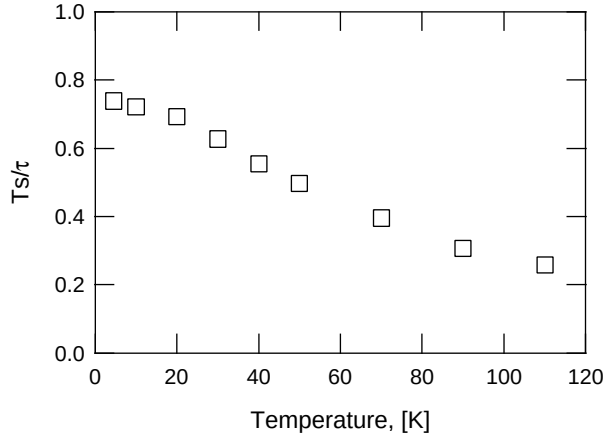


Figure 6.6: The measured values of T_S/τ as function of temperature.

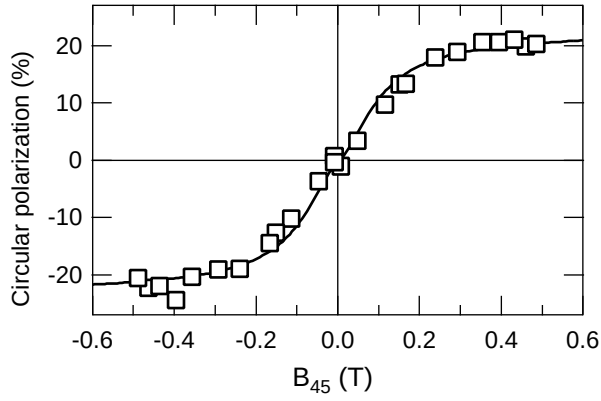


Figure 6.7: The circular polarization of the electroluminescence of the (Ga,Mn)As spin-LED at 4.6 K as function of the external magnetic field ($\square$) and the fit based on the OHE-theory (full line).

be $47 \pm 5\%$, which is considerably lower than for the annealed sample. In Fig. 6.9(b) the temperature dependence of the injected spin polarization is shown. The Curie temperature is about 60 K, consistent with other as-grown (Ga,Mn)As samples, grown with similar conditions.

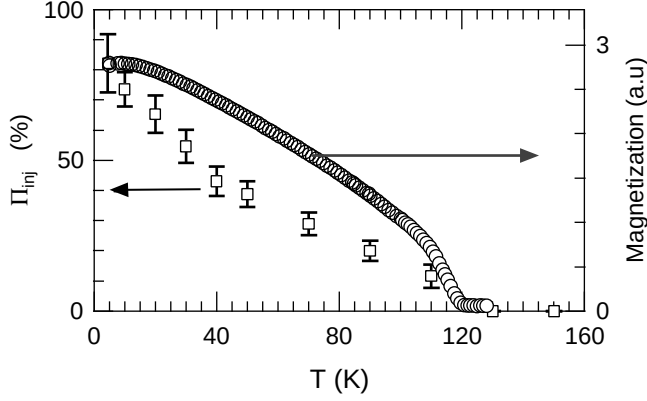


Figure 6.8: The temperature dependence of the injected spin polarization ($\square$) and the remanent in-plane magnetization ($\circ$) of the (Ga,Mn)As film.

In Fig 6.10 the typical behaviour of the degree of injected spin polarization on the applied bias voltage (a) and the current (b) is shown at 4.6 K. The spin polarization decreases dramatically at higher bias to vanish almost completely above 2.5 V.

6.3 The bias dependence of the measured spin polarization.

It is striking that the spin polarization completely vanishes at higher applied biases. For the prospects of using these results for the design and fabrication of novel spintronic devices, a detailed understanding of spin polarized Zener tunneling in (Ga,Mn)As-based semiconductor heterostructures is of primary importance. We have theoretically analyzed the transport in the spin-LED structures as a function of the applied bias by two different approaches, each of them particularly appropriate to elucidate specific issues which occur in this problem. First, we model the charge transport in the entire LED under bias using Medici. By these simulations a detailed understanding of the charge distribution, band bending, electron and hole currents and their bias dependence in the entire device can be achieved. However, to explain the observed very high injected spin polarization a more detailed description of the spin-dependent band structure in the heterostructure is needed. This part of the theoretical treatment of the problem has been done in close collaboration with the institute of physics (IFPAN) in War-

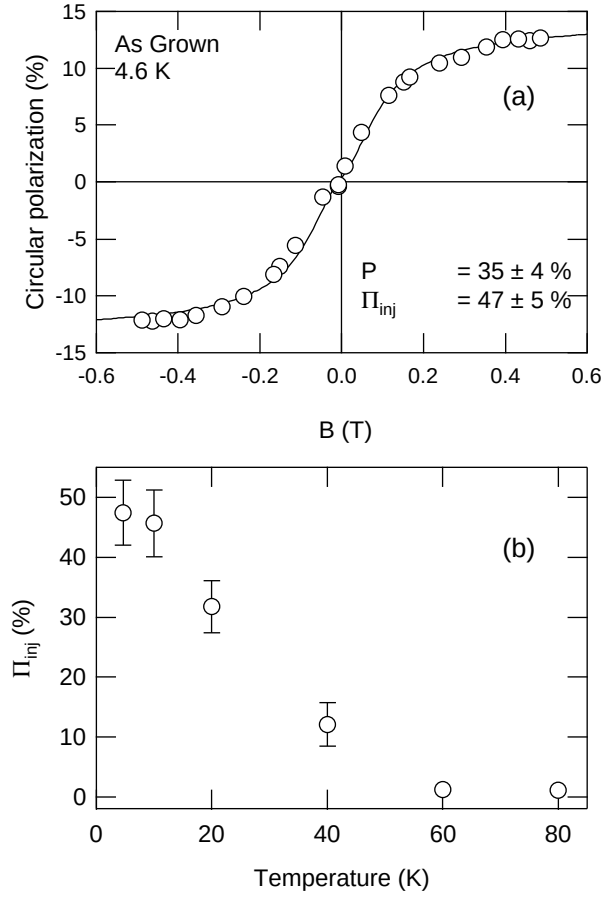


Figure 6.9: (a) The circular polarization of the electroluminescence of the (Ga,Mn)As spin-LED as function of the external magnetic field and the fit based on the OHE theory (full line). The (Ga,Mn)As injector has not undergone an annealing step. (b) The temperature dependence of the fitted injected spin polarization.

saw, Poland. The calculations were mainly performed in Warsaw, based on the band profiles taken from the self-consistent simulations, performed in Leuven, while the interpretation of the results of the calculations was a common achievement. In order to study the current spin polarization, a model of interband tunneling has been generalized, which combines an em-

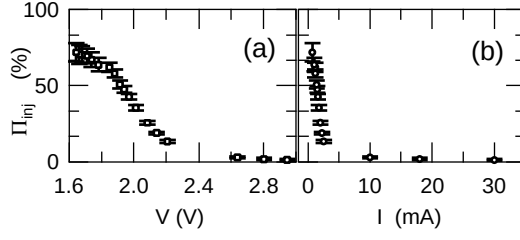


Figure 6.10: The degree of injected spin polarization measured at 4.6 K as a function of the applied bias voltage (a) and the current (b).

pirical tight-binding approach with the Landauer-Büttiker formalism [9] to the case of coherent spin transport. Importantly, in contrast to the standard $k \cdot p$ method, the scattering formalism based on the tight-binding scheme takes into account all the effects resulting from the electric field in the depletion zone of the diode, in particular the Rashba and Dresselhaus terms, essential for the tunneling.[10] The cost of these advantages comes with the reduction of the possible size of the simulated device. Hence, the whole LED structure can not be considered by this approach. Thus, in the calculation of the current spin polarization we concentrate only on the description of the (Ga,Mn)As/GaAs interface, however, using the interface band profiles obtained by the self-consistent Medici simulations of the entire device.

6.3.1 Electron and hole currents

In the self-consistent Medici simulations the (Ga,Mn)As region is treated as a heavily doped GaAs region. The value of the doping level is chosen based on transport measurements on samples grown using similar growth conditions on a semi-insulating substrate. The interband tunneling is taken into account using the Kane model[11] (following the $k \cdot p$ method) to introduce a generation term.[12] We compute the generation of carriers due to interband tunneling assuming a completely filled valence band. Due to the large Fermi energy (E_F) in (Ga,Mn)As, the carriers depart from E_F below the valence band maximum. This results in a shift of the threshold voltage where interband tunneling starts, but does not qualitatively alter the results of the calculations. The estimated size of this error, for the assumed hole concentration of $3.5 \times 10^{20} \text{ cm}^{-3}$, is about 0.25 V. The impact ionization is treated in a post-processing mode, *i.e.*, the carrier generation is calculated after the self-consistent calculation of the band bending. Due to well-known numerical convergence issues, it was not possible to simulate the structure

at very low temperatures, at which the experiment was performed.

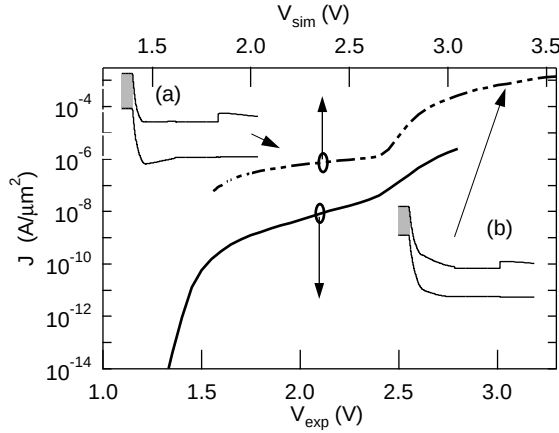


Figure 6.11: The measured current at 4.6 K (solid line) and the simulated current for 40 K (dashed line) as a function of the applied bias voltage. The voltage axis of the simulated curve has been shifted by 0.25 V to take into account the offset in the onset of tunneling. The insets show the simulated conduction and valence band profiles at (a) 1.81 V and (b) 3.0 V bias. The grey-colored area represents the (Ga,Mn)As injector.

In Fig. 6.11 the simulated current density at 40 K is shown together with the measured current density at 4.6 K as a function of the applied bias voltage. Taking into account all the above mentioned discrepancies and the fact that the doping levels and aluminum concentrations used in the simulations are nominal and can differ from real ones, the presented comparison between simulated and the measured behavior shows a good qualitative agreement.

Between 1.6 V and 2.3-2.5 V the measured current is dominated by interband tunneling of electrons through the (Ga,Mn)As/GaAs tunnel diode, while above 2.6 V the current is dominated by the holes, which can freely flow from the substrate to the top contact. Figure 6.12 shows the band diagram (solid lines) of the (Ga,Mn)As/(Al,Ga)As diode together with the Al concentration and the carrier generation due to interband tunneling (dark filled) as a function of the depth near the (Ga,Mn)As/GaAs interface at 1.81 V and at 3 V. There are two peaks present in the plotted carrier generation, one in the (Ga,Mn)As area, which shows the generated holes in the (Ga,Mn)As valence band, and one in the GaAs area that shows the

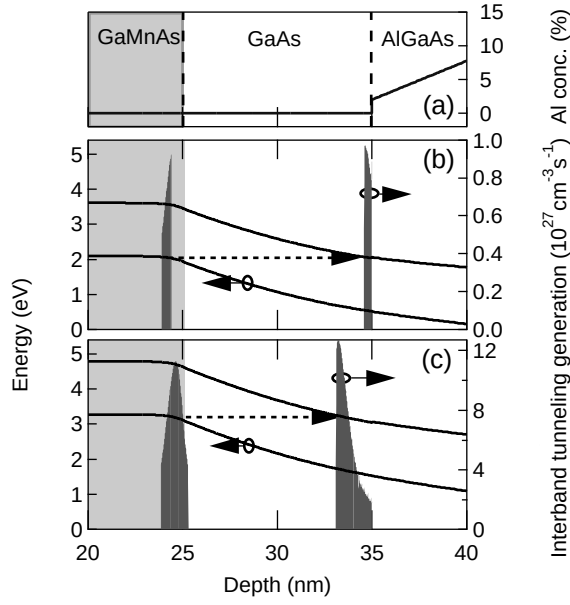


Figure 6.12: The Al concentration as a function of the depth near the (Ga,Mn)As/GaAs tunnel diode (a). The band diagram (solid lines) and the carrier generation (dark filled) due to interband tunneling at 1.81 V (b) and 3 V bias voltage (c). The light filled areas represent the (Ga,Mn)As injector. The dotted arrows indicate the tunneling of valence electrons from (Ga,Mn)As into the conduction band of GaAs.

generated electrons in the GaAs conduction band. From this picture we can deduce that interband tunneling takes place from the valence band of (Ga,Mn)As to the conduction band of GaAs. When GaAs changes into (Al,Ga)As, at a depth of 35 nm, the increasing Al concentration causes a widening of the band gap and an abrupt increase of the tunneling distance. This results in an exponentially smaller tunneling probability and hence the number of electrons that is generated in the (Al,Ga)As region is negligible. This means that only the part of the (Ga,Mn)As valence band that overlaps with the GaAs conduction band can participate in the tunneling process. If we compare the low and the high bias cases we see that the area from where electrons tunnel (the left peak in the carrier generation) is much bigger at high bias than at low bias. When the voltage drop over the tunnel diode increases, the part of the valence band of (Ga,Mn)As that overlaps with the conduction band of GaAs increases and hence the tunneling originates

from a wider region and a larger energy window in the (Ga,Mn)As valence band. In Fig. 6.13 we show the hole concentration and the (normalized)

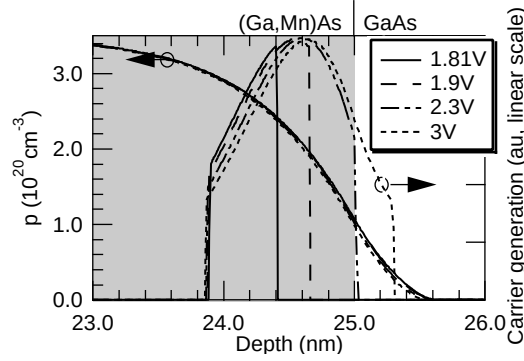


Figure 6.13: The hole concentration (left) and the carrier generation due to interband tunneling (right) at different values of the applied bias voltage near the (Ga,Mn)As/GaAs interface. The grey-coloured area represents the (Ga,Mn)As injector.

carrier generation due to interband tunneling as a function of the depth near the (Ga,Mn)As/GaAs interface for different values of the applied bias voltage. The simulations show that the hole concentration decreases in the bottom two nanometer of the (Ga,Mn)As layer due to the band bending in the (Ga,Mn)As/GaAs p-n-diode. Increasing the voltage has a negligible influence on the hole depletion near the interface. However, we do see a big change in the properties of the tunneling region. At low bias, the tunneling electrons mainly depart from a region where the hole concentration is only slightly smaller than the bulk value of $3.5 \times 10^{20} \text{ cm}^{-3}$, while at higher bias this region shifts and widens and electrons can tunnel from a region where the hole concentration is much lower. At a somewhat higher bias ($> 2.2 \text{ V}$) we see that electrons also tunnel out of the valence band of the non-magnetic GaAs layer. In the experiment this bias will probably be lower due to the above described error in the interband tunneling model.

Apart from Zener tunneling, also impact ionization can contribute to the generation of electrons in the GaAs conduction band. This process generates unpolarized electron-hole pairs and hence dilutes the injected spins and diminishes the measured spin polarization. Impact ionization in this case is caused by holes that flow from the substrate to the top contact and are heavily accelerated by the strong electric field near the (Ga,Mn)As/GaAs interface. Fig. 6.14 shows the comparison of the carrier generation induced by interband tunneling and impact ionization at different values of the applied

bias. This indicates that the carrier generation due to impact ionization only starts dominating at the second hump in the IV-characteristics, where the valence band reaches a flat-band situation (Fig. 6.11, inset (b)) and the holes can flow freely from the substrate to the top contact. Below this "hump" impact ionization can be neglected.

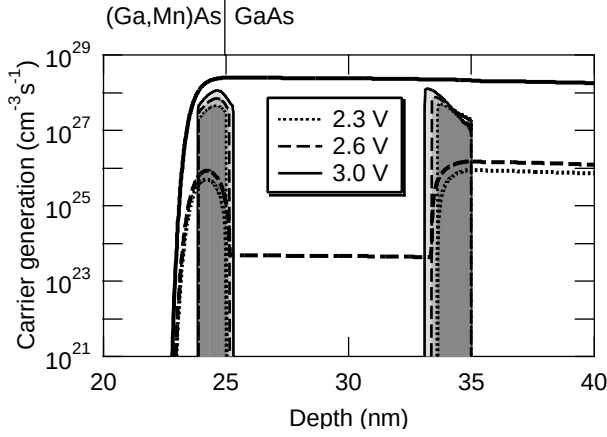


Figure 6.14: The carrier generation due to impact ionization and to inter-band tunneling at different values of the applied bias voltage. The grey-shaded areas represent the carrier generation due to interband tunneling in the (Ga,Mn)As valence band and the GaAs conduction band.

6.3.2 Current spin polarization

The self-consistent Medici simulations of the device provide us with the band profiles that are further used in the calculation of the current spin polarization. Since the ballistic tunneling of electrons from the valence band of (Ga,Mn)As to the conduction band of GaAs is essential to understand the experimental data, the Landauer-Büttiker formalism seems to be a method of choice. Thus, to describe the spin effects, we implement the spin dependent tight-binding Hamiltonian into the Landauer-Büttiker formalism for the coherent tunneling. These calculations have been performed by Piotr Sankowski in the institute of theoretical physics in Warsaw.[13] The transmission matrix is determined in terms of the extended transfer-matrix method within the tight-binding framework. The tight-binding Hamiltonian matrix is composed of three parts that describe the left and right leads, and the middle region, where the incoming Bloch waves from the left lead are

scattered into outgoing Bloch states of the right lead. The self-consistently calculated local electrostatic potential as well as band offsets are incorporated to the orbital "on site" energies. [9] Well established numerical techniques allow us to determine the transfer coefficients $t_{k_{\perp,i} \rightarrow k_{\perp,j}}(E, \mathbf{k}_{\parallel})$, which describe the probability of tunneling from the incoming state $k_{\perp,i}$ to the outgoing state $k_{\perp,j}$ for a given electron energy E and a wave vector parallel to the surface $\mathbf{k}_{\parallel}$. [14] The tunneling current $\mathbf{j}$ takes the form:

$$\mathbf{j} = \frac{-e}{4\pi^3\hbar} \int_{BZ} d^2k_{\parallel} dE \sum_{\substack{k_{\perp,i}, k_{\perp,j} \\ v_{\perp,i}, v_{\perp,j} > 0}} [f_L(E) - f_R(E)] |t_{k_{\perp,i} \rightarrow k_{\perp,j}}(E, \mathbf{k}_{\parallel})|^2 \frac{v_{\perp,j}}{v_{\perp,i}}, \quad (6.1)$$

where f_L and f_R are the electron Fermi distributions in the left and right interface and $v_{\perp,i}$ are the group velocities of the corresponding Bloch states. To determine the current spin polarization we use the above formula to compute separately the currents of spin up and spin down electrons. The spin up current $\mathbf{j}^{\uparrow}$ is given by Eq. (6.1), where we sum only over spin up (determined by the direction of magnetization in the ferromagnetic material) outgoing states $k_{\perp,j}^{\uparrow}$. The spin down current $\mathbf{j}^{\downarrow}$ can be obtained in a similar way. The current spin polarization P_j is defined as follows:

$$P_j = \frac{\mathbf{j}^{\uparrow} - \mathbf{j}^{\downarrow}}{\mathbf{j}^{\uparrow} + \mathbf{j}^{\downarrow}}.$$

In order to construct the empirical tight-binding Hamiltonian matrix for the heterostructure we have to start from the description of the constituent materials. To describe the band structure of the bulk GaAs we use the $sp^3d^5s^*$ tight-binding parametrization, with the spin-orbit coupling included, as proposed by Jancu *et al.* [15] This model reproduces correctly the effective masses and the band structure of GaAs in the whole Brillouin zone, in agreement with the results obtained by empirically corrected pseudopotential method. The parametrization includes only the nearest neighbor (NN) interactions. For each anion and cation 20 orbitals are used - hence, with each GaAs layer (0.28 nm) of the structure the size of the tight-binding matrix increases by 40. The same set of orbitals is used to describe the (Ga,Mn)As layer. It should be pointed out that the d orbitals used in our $sp^3d^5s^*$ parametrization are not related to the Mn ions incorporated into GaAs. The presence of Mn ions in (Ga,Mn)As is taken into account only by including the $sp-d$ exchange interactions within the virtual crystal and mean-field approximations. The values of the exchange constants are determined by the experimentally obtained spin splittings: $N_0\alpha = 0.2$ eV for the conduction band and $N_0\beta = -1.2$ eV for the valence band. [16] The

other parameters of the model for the (Ga,Mn)As material are taken to be the same as for GaAs – this is well motivated because the valence-band structure of (Ga,Mn)As with small fraction of Mn has been shown to be quite similar to that of GaAs. [16] We construct the tight binding matrix for the heterostructure taking for each two (cation + anion) monolayers of the structure the description of the corresponding bulk material. The NN interactions between GaAs and (Ga,Mn)As are described by the same parameters as the interactions in bulk GaAs. Consequently, the valence band offset between (Ga,Mn)As and GaAs originates only from the spin splitting of the bands in (Ga,Mn)As.

The Fermi energy in the included materials is determined by summing up the occupied states over the entire Brillouin zone. The number of occupied states is determined by the assumed carrier concentration in the material. The calculations of the Fermi energy dependence versus hole concentration are consistent with the corresponding results presented in Ref. [17]. To get a physical insight into mechanisms leading to the high spin current polarization, we consider the Zener diode region in the spin-LED device, i.e., the p -Ga_{1-x}Mn_xAs/ n -GaAs heterojunction. While in such a model the bias voltage is only applied over the Zener diode, in contrast to the actual device, it should provide correct information on the spin polarization. In our computation, we assume that the magnetization vector is at 27° out of plane, as implied by experimental conditions. We evaluate the spin current polarization P_j with respect to this direction.

According to the self-consistent calculations, the scattering region consists of approximately 120 monolayers of (Ga,Mn)As and GaAs, and corresponds roughly to the depletion zone of the diode. These results are employed in order to describe the potential profile in the tight-binding calculations, as shown in Fig. 6.16(a). In particular, the self-consistently calculated electrostatic potential that describes the band bending for $V_0 = 1.81$ V, i.e., for the voltage when the interband tunneling process starts, is the basis for the potential profile that is incorporated into the tight-binding matrix. An additional component of the potential profile is the bias ΔV that corresponds to the potential drop across the Zener diode when the bias on the whole spin-LED is changed. We assume a linear potential drop which fairly well renders the real band bending across the p - n -junction.

Figure 6.16 shows P_j as a function of the bias ΔV in the Zener tunneling diode. We see that at low bias, P_j is of the order of 0.6, in good agreement with the experimental results (0.7-0.8). Interestingly, P_j decreases rapidly with ΔV . The strong dependence of P_j on bias is again in agreement with the experimental findings, though a direct comparison is hampered by the fact that the exact relation between ΔV and the total bias V applied to the device is known only from simulations.

Having these obstacles in mind and to reduce computational burden,

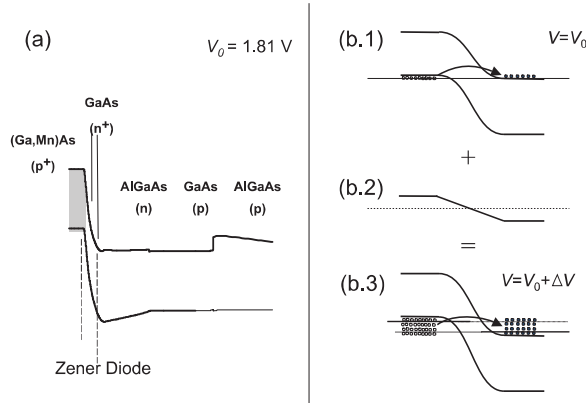


Figure 6.15: Scheme of the structure description used in the calculations: (a) the band profile of the entire LED calculated using Medici for $V_0 = 1.81 \text{ V}$; (b.1) in the tight-binding calculations only the Zener diode region is considered; (b.2) a linear band bending given by an applied bias ΔV is added; (b.3) the band profile used in the tight-binding calculations is a sum of (b.1) and (b.2).

the calculations for higher bias ($\Delta V > 0.1 \text{ V}$) have been performed with lower accuracy – the latter is probably responsible for the spread of obtained polarizations in this region.

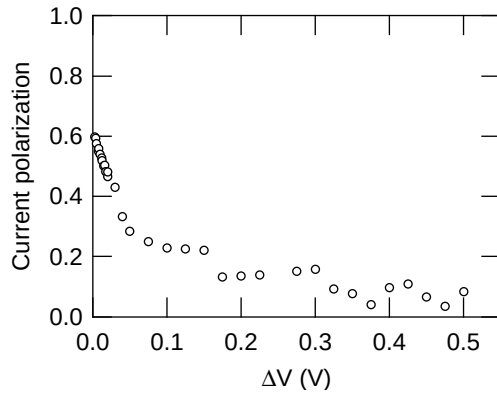


Figure 6.16: The calculated bias dependence of the current polarization for the structure in Fig. 6.15(b).

6.3.3 Discussion

Both theoretical approaches point towards a decrease of the injected spin polarization with bias. It is clear from the self-consistent Medici simulations that increasing the bias leads to an enhanced tunneling from strongly depleted (Ga,Mn)As and non-magnetic GaAs and to an increase of the energy window from where tunneling occurs. What exactly happens in the depleted region remains unclear at this stage. The decrease of the Fermi energy in the depleted (Ga,Mn)As seems to indicate a higher spin polarization at the Fermi energy in this region of the device. It is, however, not exactly known how the exchange splitting of the valence bands behaves in the regime of strong depletion. Also the exchange splitting may be reduced in this regime, causing a decrease of the spin polarization.[20] At higher bias, a significant part of the tunneling current will originate from the valence band of non-magnetic GaAs, where there is no exchange-splitting of the bands and hence the spin polarization is zero.

The results of the simple tight-binding model of coherent tunneling, which describes carefully the band structure of the heterostructure, especially the interface region, agree well with the experimental data, both regarding the maximum injected spin polarization and the bias dependence of injected spin polarization. This fact suggests that here the decrease of the spin-polarization of the tunneling current with the applied dc bias has a more fundamental origin, related to band structure effects.

The strong dependence of P_j on ΔV can be related to the fact that the current polarization is determined not only by the electron spin polarization at the Fermi energy, but also by the transmission coefficients through the structure for electrons below the Fermi energy in (Ga,Mn)As and to higher states in the conduction band of GaAs, as shown schematically in Fig. 6.15. When we increase the bias we start to have an increased contribution to the tunneling current from the minority spin subbands. An important aspect of the coherent transport through a structure is the "group velocity mismatch". Namely, due to the conservation of the transverse quasi-momentum and of the energy, the transmission coefficient through an interface decreases with the difference between the group velocities in the two materials at the energy in question, i.e., (Ga,Mn)As and undoped GaAs spacer. The "velocity mismatch" can result in a smaller transmission coefficient through the GaAs spacer for the majority spin electrons as compared to the minority spin electrons. The minority spin electrons have a smaller quasi-momentum and hence their velocities match better. Thus, one can argue that the bias, by increasing the concentration of the minority spin electrons and by increasing the importance of the region with a small group velocity, enhances the tunneling of minority spin electrons, which reduces the current spin polarization with bias seen in Fig. 6.16.

In order to understand the tunneling process better simplified calculations have been performed in Warsaw for the (Ga,Mn)As/GaAs p - n -diode in which we restrict the scattering region to few layers, as shown on Fig. 6.17. Guided by previous theories of tunneling magnetoresistance (TMR), we expect that while such a model overestimates necessarily the tunneling current, it can provide quantitative information on current spin polarization that is determined by inter-band tunneling matrix elements and degree of spin polarization in the ferromagnetic electrode.



Figure 6.17: Scheme of the (abrupt) (Ga,Mn)As/GaAs p - n -diode structure used in the simplified tight-binding calculations at zero and reverse bias.

In Fig. 6.18 the dependence of the tunneling current and its spin polarization on the k -vector in the (Ga,Mn)As valence sub-bands are presented. These results show that in the tunneling process practically only the light holes take part. This can be caused by a better matching of their transverse k vectors with the transverse k vectors of the conduction band states in the n -GaAs. Secondly, in the interband tunneling process, the tunneling electrons travel on an imaginary band while traversing the forbidden energy zone. As can be seen in Fig. 6.19, in GaAs only the light hole band connects to the first conduction band while the heavy hole band connects to a higher energy conduction band.[9] This is, however, only true for tunneling from states at the Γ -point. Due to the large Fermi energy, the tunneling electrons all possess a significant $k_{||}$, which allows also the tunneling of electrons from the heavy-hole bands. In Fig. 6.18(b) one may also note that at the Fermi level for the considered hole concentration, the contribution to the tunneling from majority states increases with k . Hence, also the current spin polarization increases with k .

Triggered by the strong anisotropy of the magnetoresistance observed in recent experiments [19], we have examined also, both theoretically and experimentally, the angular dependence of the injected spin polarization. In the case of Zener tunneling, however, only a small, ca 6%, difference between [110] and [1-10] directions has been both observed and obtained in the calculations. This result agrees with the observation that the light holes – which have a smaller anisotropy in k -space, as suggested by Fig. 6.18 – participate

considerably in the interband Zener tunneling in the (Ga,Mn)As/n-GaAs diode.

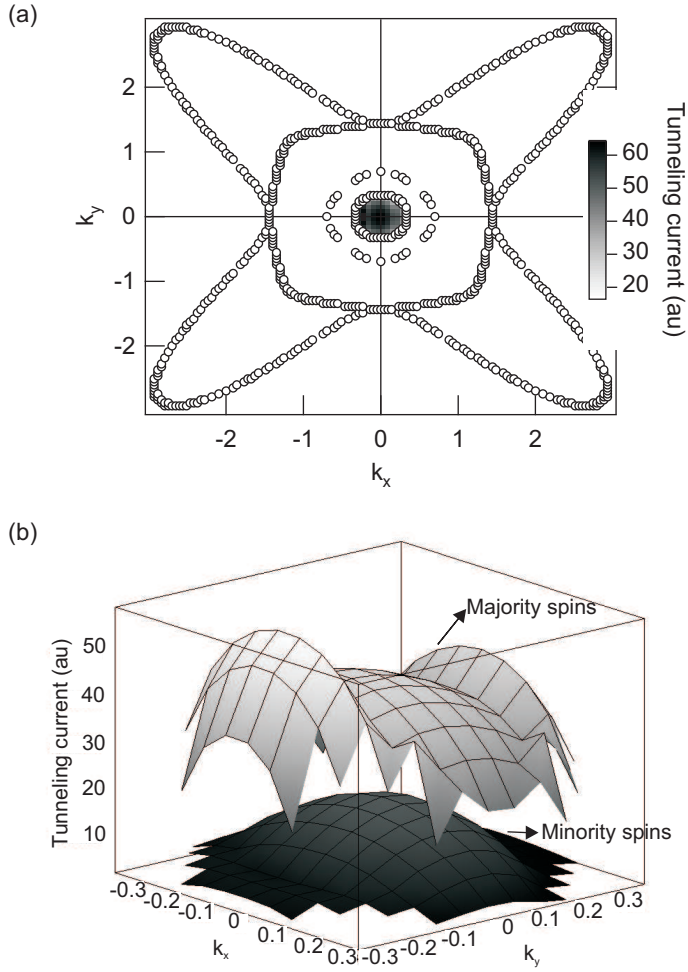


Figure 6.18: a) The calculated valence band intersection of (Ga,Mn)As for a hole concentration of $3.5 \times 10^{20} \text{cm}^{-3}$ and the tunneling current as function of k_x and k_y . b) The magnitude of the tunneling currents for both the majority spins and the minority spins for different $k_{\parallel}$ -vectors in the structure presented in Fig. 6.17 for zero bias.

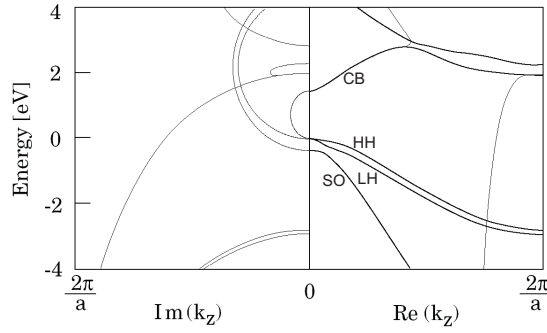


Figure 6.19: The complex band structure of GaAs, adopted from Ref. [9]. The light hole band connects to the lower conduction band, while the heavy-hole band connects to a higher energy conduction band

6.4 The effects of the measurement temperature and annealing

As is depicted in Fig. 6.8, the measured injected spin polarization behaves qualitatively different from the bulk magnetization as function of the temperature, although the critical temperature is the same. This agrees with the findings of Chun *et al*[21] who have measured the TMR of a MnAs/AlAs/(Ga,Mn)As magnetic tunnel junction as function of the temperature and found that indeed the TMR follows a different behaviour with respect to the temperature dependence of the (Ga,Mn)As magnetization. They ascribe this discrepancy to a possible faster decay of surface magnetism or to the possibility that the spin polarization in (Ga,Mn)As is not directly proportional to the magnetization. In our case, the depletion of the (Ga,Mn)As/GaAs- interface and the corresponding quenching of the hole concentration can lead to a gradual change of the Curie temperature of the interfacial (Ga,Mn)As. Since tunneling is an interfacial effect, this may lead to a deviation of the behaviour of the bulk magnetization. Furthermore, the Zener-spin-LED consists of two diodes in series. The resistance of the bottom diode (substrate/n-AlGaAs) drops exponentially with temperature, in contrast to the tunnel diode, which has a weak temperature dependence. This means that at constant bias voltage and increasing temperature, the voltage drop over the Zener diode increases. Since the injected spin polarization strongly decreases as a function of the applied bias, the injected spin polarization can drop faster with temperature than the magnetization.

The injected spin polarization of the non-annealed sample, as depicted in Fig. 6.9 is significantly smaller than the polarization of the annealed sample. The relatively high Mn-concentration (8%) in the as-grown sample gives rise to a high degree of compensation due to Mn-interstitials. The interstitials, being double donors, cause a decrease of the hole concentration, lowering the Fermi energy. Although a mere decrease of the Fermi energy would rather indicate an increase of the spin polarization, this is only true for a fixed magnetization (which depends on the effective Mn-concentration). However, the Mn interstitials also cause a decrease of the magnetically active substitutional Mn. The interstitial Mn atoms prefer to be located close to a substitutional Mn atom and couple strongly antiferromagnetically with the substitutional Mn, reducing the amount of magnetically active Mn.[22] Hence, the strong reduction of both the magnetization and the hole concentration can give rise to a smaller spin polarization, as experimentally observed.

6.5 Conclusions

In conclusion, we have demonstrated highly efficient spin injection from a (Ga,Mn)As top injector into an AlGaAs/GaAs spin-LED using an interband tunneling process. The injected polarization vanishes at 120 K, the Curie temperature of the (Ga,Mn)As layer and exhibits a strong dependence on the applied bias. We have analyzed the spin polarization of the injected current in a (Ga,Mn)As/(Al,Ga)As spin-LED by performing self-consistent simulations of the band bending of the device under bias. Furthermore, in a collaboration with a theoretical group in Warsaw, we have investigated the spin polarization of the tunneling current using a tight-binding model together with the Landauer-Büttiker formalism. Our studies explain quantitatively the large spin polarization of the injected current and its strong dependence on the applied voltage. Our Medici simulations point to an enhanced tunneling from the depleted (Ga,Mn)As and non-magnetic GaAs regions and an increase of the energy window where tunneling is allowed. Moreover, the results of the tight-binding calculations of coherent tunneling indicate that the spin polarization decreases with the applied voltage because the enhanced energy window of tunneling at higher bias leads to an increase of the relative contribution of the minority spin electrons in the tunnel current due to the better match of their group velocity with the group velocity of the conduction band states.

The published results presented in this Chapter have recently been confirmed by Kohda *et al*[23], who obtained similar results while adopting the semiconductor structure as used in this work.

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Chapter 7

The electrical properties of the different spin-LEDs.

In the previous three Chapters three types of spin-injector structures (oxide based tunnel injectors, Schottky-barrier tunnel injectors and (Ga,Mn)As based interband-tunnel injectors) have been fabricated and examined. We have analyzed their operation in terms of the absolute injected spin polarization and the bias dependence of the measured spin polarization. In this short Chapter we compare the different injector structures with respect to their electrical properties, which have proved to differ significantly.

Especially the oxide injectors behave quite different from the two other injector structures, namely the Schottky-barrier injectors and the interband-tunneling injectors, that show a comparable behaviour.

A final issue we discuss in this Chapter is the effect of impact ionization. Under high bias conditions, this effect can create unpolarized electron-hole pairs, diluting the injected spins.

7.1 The oxide injectors

In spin-LEDs we need a large concentration of both electrons and holes in the active region. Electrons are injected from the magnetic top contact, while holes are supplied by the p-type substrate. When using an oxide insulator as a tunnel barrier in the injector, the interface states at the (Al,Ga)As-oxide interface pin the Fermi level just below halfway the (Al,Ga)As energy gap. The Fermi-level pinning results in the formation of a Schottky barrier and a depleted region in the semiconductor. This causes the energy difference between the top metal contact and the (Al,Ga)As conduction band to be about 0.8 eV. Since the electrons need to tunnel

in the conduction band of the (Al,Ga)As top region in order to be collected in the GaAs active region afterwards, this energy difference needs to be accommodated by the oxide tunnel barrier. The associated voltage drop implies the existence of negative charges at the metal side and positive charges at the semiconductor side. The high carrier concentration in the metal can easily supply the necessary negative charges while in the semiconductor the positive charges must be supplied by holes that accumulate at the oxide-semiconductor interface. The existence of the depleted Schottky barrier prohibits hole accumulation at low bias, but upon application of a bias voltage, the induced band bending allows holes to flow from the substrate to the top contact. We have supported this picture by self-consistent simulations.

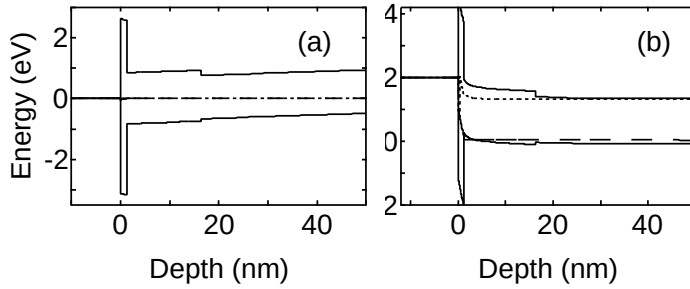


Figure 7.1: The band diagram of a Metal/Oxide/(Al)GaAs LED near the metal/oxide-interface at zero bias (a) and under an applied bias of 2 V (b). The dotted and the dashed lines represent the quasi-Fermi levels for electrons and holes, respectively. Near the oxide/AlGaAs-interface the presence of a large hole accumulation is clear from the relative positions of the hole quasi-Fermi level and the valence band.

We consider here the case of a CoFe-AlO_x-AlGaAs (undoped)- GaAs (undoped)-AlGaAs (p-type)-GaAs (p-type) structure. Upon the application of an external bias voltage, the initial voltage drop takes place over the semiconductor part of the Schottky diode. When reaching flatband conditions in the semiconductor (near 0.7 V), holes start to accumulate at the AlGaAs/AlO_x-interface and provide the required positive charge to allow a voltage drop over the oxide. Near 1.6 V the metal Fermi level then reaches the conduction band of AlGaAs and spin-polarized electrons start to tunnel from the metal into the conduction band of AlGaAs. In Fig. 7.1 the band diagram for this device is shown at zero bias and under an applied bias of 2 V. At zero bias, the Schottky diode causes a hole depletion near the

oxide/AlGaAs interface, while under bias at the interface of the oxide and AlGaAs a clear hole accumulation is observed.

The need for the hole accumulation at the oxide-semiconductor interface causes the existence of a high hole current at the luminescence threshold of the spin-LED. The observed high luminescence threshold and operation currents support this picture. A typical threshold current is about 400 A/cm^{-2} at 80 K.

7.2 The Schottky- and interband-tunneling injectors

The Schottky tunnel injectors and the interband tunnel injectors operate in a quite different fashion compared to the oxide injectors. In the case of the Schottky tunnel barrier the energy difference between the top metal contact and the (Al,Ga)As conduction band must be accommodated by the built-in voltage drop of the Schottky barrier. By tailoring the doping level this built-in voltage drop equals the height of the Schottky barrier. Hence, electrons can tunnel already at zero bias from the metal contact into the (Al,Ga)As conduction band. A similar situation occurs in the case of the interband-tunneling injectors, where the barrier height is given by the bandgap. In both cases, the electron and hole currents are much more balanced, resulting in much lower threshold currents, in the order of 0.5 to 5 A/cm^{-2} at 80 K, which represents a significant improvement with respect to the oxide injectors. Counterintuitively, the lack of a tunnel barrier for the holes (as in the case of the oxide injectors) does not lead necessarily to high hole currents. In a flatband situation the electron and the hole current contribute almost equal to the total current. The surface electric fields that might attract holes are screened by the high doping level near the surface. Furthermore, by engineering the Al concentration in the spin-transport region, as in the case of the (Ga,Mn)As based interband-tunneling injectors, one can create a barrier for the holes, while the electrons see a flatband situation. This has been shown already in Fig. 6.16.

Specifically in our structures, the balanced electron and hole currents allow much larger electron currents than in the case of the oxide injectors. The resulting high electron concentration in the active region is then responsible for a large electron-nuclear interaction, as will be shown in Chapter 8.

7.3 Additional, parasitic sources of unpolarized currents: Impact ionization

Under high bias conditions a source of unpolarized electrons can arise inside the LED structures. Impact ionization at sufficient bias can lead to the creation of unpolarized electron-hole pairs, leading to a dilution of the injected spins. Especially in the case of the Schottky- and the interband tunneling injectors, where high electric fields are present near the metal contact, impact ionization can occur. Indeed, in the previous Chapter, simulations indicated that above 2.6 V, i.e. above flatband conditions the hole current caused a strong contribution of impact ionization to the carrier generation in the device. This has been shown in Fig. 6.14.

Furthermore, impact ionization can lead to luminescence in situations where there is no or very little injection of electrons through the contact present, creating the necessary electron-hole pairs in the active region. In early experiments we have studied NiMnSb injectors without heavily doping the semiconductor region near the interface. Although the devices showed luminescence, the emitted light was completely unpolarized. Electrical measurements confirmed that the Fermi level was pinned 0.8 eV below the conduction band edge, and simulations showed that under these conditions no electrons are injected into the semiconductor (Fig. 7.2(a)). However, at sufficiently large bias, impact ionization by the hole current can generate unpolarized electron-hole pairs. When this effect is taken into account, the simulations show a large electron concentration in the active region (Fig. 7.2(b)), which was responsible for the unpolarized luminescence.

7.4 Conclusion

In this Chapter we have discussed the different electrical properties of the different injector contacts, focussing on the different internal device currents, electron current, hole current and currents caused by impact ionization. This leads to a different behaviour for the oxide injectors compared to the Schottky-injectors and the interband-tunneling injectors. Furthermore, one should be careful by measuring the luminescence as a measure for the injected spin polarization as impact ionization can add unpolarized electron-hole pairs to the electrically injected electrons.

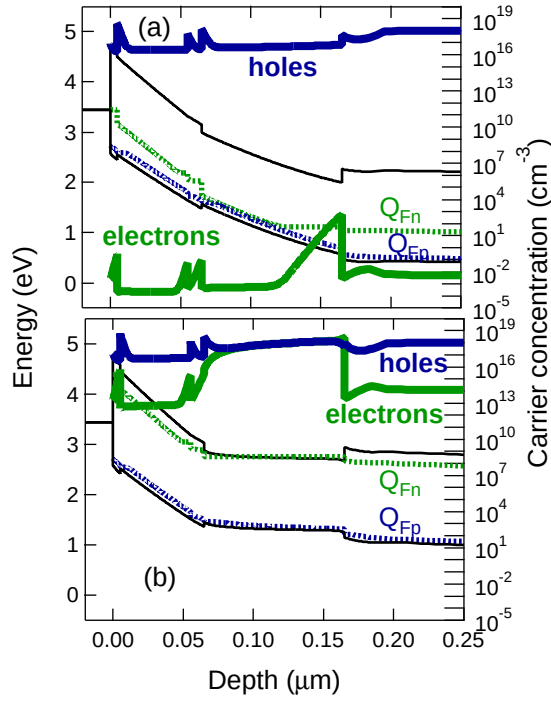


Figure 7.2: The band diagram of a Metal/(Al)GaAs LED without heavy doping near the metal/semiconductor interface at an applied voltage of 3.5 V, (a) without taking impact ionization into account and (b) including of impact ionization in the simulations. The dotted green and blue curves corresponds to the quasi-Fermi levels of the electrons and the holes, respectively. The full green and blue curves correspond to the electron and hole concentrations, respectively.

Chapter 8

Dynamic Nuclear polarization

8.1 Introduction

It is known that spin-polarized electrons in the conduction band of GaAs can cool the the nuclear spin system by polarizing the nuclear spins in the GaAs lattice by the hyperfine interaction. The magnitude of this polarization depends on the localization of the electron wave function and the amount of electrons that are involved. In early experiments, a large nuclear spin polarization due to optical pumping has been detected in compensated samples, where electron localization on donor sites takes place.[1] In recent experiments electrical spin injection from an Fe contact in a GaAs quantum well resulted in a steady state nuclear polarization.[2]

In this chapter, we discuss the effects of electrical spin injection from a MnSb contact into an AlGaAs/GaAs-based spin light emitting diode (LED) on the polarization of the nuclei in the GaAs bulk active region. The uniaxial anisotropy and the high coercivity of the MnSb ferromagnetic film allow us to control the sign and the magnitude of the injected spins and the magnetic field independently. We demonstrate real-time transient effects in the electronic spin polarization that can be associated with a slow change of the nuclear polarization when the sign of the injected spins is reversed. Furthermore we show that a reversal of the magnetic field results in an adiabatic reversal of the nuclear spin polarization. Using these transient effects, we were able to measure the nuclear spin relaxation time due to both the hyperfine interaction and the relaxation time in the absence of electrons. Finally, to further investigate the behavior of the nuclei, we bring the nuclear spins into resonance using an oscillating magnetic field and look

at the effect on the electron spins. From the nuclear magnetic resonance measurements and the field and bias dependence of the nuclear relaxation times we explain the magnitude and the bias dependence of the low-field nuclear depolarization.

8.2 Experiments.

We have studied dynamical nuclear polarization in the Schottky spin-LEDs from Chapter 5. In these devices spin injection has been realized from epitaxial MnSb layers into GaAs through a Schottky-barrier. Through the Schottky barriers, larger electron injection currents can be achieved than through oxide barriers, which is important for nuclear polarization.[4; 5] Figure 8.1 shows the simulated band structure of the spin-LED under bias.[6] The band structure and the doping profile of the LED were designed such that under bias the spin-polarized electrons tunnel through the highly n-type doped (Al,Ga)As interfacial region and then travel through a lightly n-type doped (Al,Ga)As region to a GaAs active region. In the GaAs-region the electrons thermalize and recombine with holes supplied from the p-type substrate.

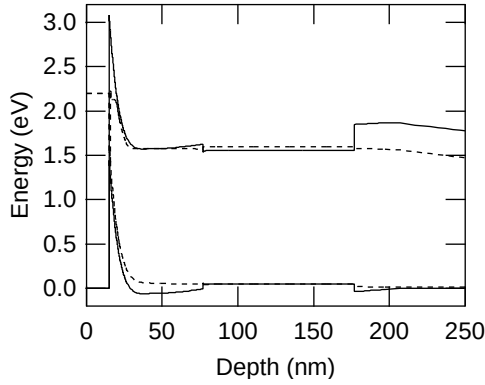


Figure 8.1: The simulated band structure of the spin-LED under bias (2.2 V) at 80 K. The dashed lines indicate the quasi-Fermi levels for electrons and holes.

MnSb is a ferromagnetic material that is particularly suited to study the effects of the magnetization on the nuclear polarization in GaAs. As shown in Fig. 5.2, it has a strong uniaxial in-plane anisotropy. It has the hexagonal NiAs crystal structure resulting in a strong magnetocrystalline anisotropy

with the c -axis as hard direction and the basal plane as easy plane. The $[1\bar{1}0]$ easy direction exhibits square loops with a relatively large coercive field, which allows us to visualize the effects of the magnetization on the Hanle curve. Furthermore, the 100% remanence allows us to manipulate the direction and the magnitude of external field independently from the sign of the injected spins (the magnetization). An out-of-plane AGFM measurement (not shown) showed a hard-axis behavior with the saturation magnetization of the film equal to $\mu_0 M_S = 0.8$ T.

8.2.1 The OHE technique: Nuclear effects.

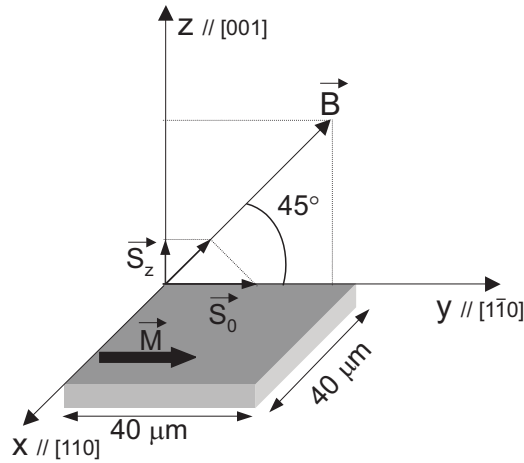


Figure 8.2: Schematic picture of the measurement geometry. The LED has been patterned in devices of 40 by 40 μm (grey area). The magnetic field is applied in an angle of 45° with respect to the plane, with its in-plane component along the easy axis of the MnSb-film. The light is collected along the sample normal.

To measure the steady state spin polarization in the GaAs active region, we analyze the degree of circular polarization of the emitted light of the (surface-emitting) spin-LED using the oblique Hanle effect technique.[8; 9] The measurement geometry is sketched in figure 8.2. The circular polarization of the emitted light is only sensitive to the perpendicular spin-component. By applying a small oblique magnetic field, with its in-plane component along the $[110]$ easy direction of MnSb, spin precession forces the average spin to come out of plane and to eventually align with the field.

In contrast to the normal Hanle effect, the magnetic field has a component parallel to the injected electron spins, which allows nuclear polarization to occur and to be observed. The injected spin-polarized electrons transfer their angular momentum to the nuclear spins through the hyperfine interaction – they pump the nuclear spin system. Again through the hyperfine interaction the polarized nuclei generate an effective magnetic field acting on the electron spins that is superimposed on the external field. This effective field can be significantly larger than the applied field. At low fields, comparable to the local dipole fields, the nuclear magnetization collapses and there occurs a sharp dip in the circular polarization.

The dependence of the average spin in the semiconductor on the external magnetic field can be calculated from the Bloch equations:[1]

$$\frac{d\mathbf{S}}{dt} = \boldsymbol{\Omega} \times \mathbf{S} - \frac{\mathbf{S}}{\tau_S} - \frac{\mathbf{S} - \mathbf{S}_0}{\tau} \quad (8.1)$$

with $\mathbf{S}$ the average spin, $\mathbf{S}_0$ the injected average spin, τ the electron-hole recombination time and τ_S the spin-relaxation time. The circular polarization is proportional to the perpendicular spin component S_Z and can in the oblique Hanle effect geometry can be calculated to be:[9]

$$P(\Omega) = \frac{\Pi_{inj} \cdot \cos(\theta)}{2} \cdot \frac{T_S}{\tau} \cdot \frac{(\Omega \cdot T_S)^2 \cdot \cos(\phi) \cdot \sin(\phi)}{1 + (\Omega \cdot T_S)^2} + \frac{\Pi_{inj} \cdot \sin(\theta)}{2} \cdot \frac{T_S}{\tau} \cdot \frac{1 + (\Omega \cdot T_S)^2 \cdot \cos^2(\phi)}{1 + (\Omega \cdot T_S)^2} \quad (8.2)$$

with Π_{inj} the degree of injected spin polarization, $T_S^{-1} = \tau^{-1} + \tau_S^{-1}$. θ is the angle between the sample plane and the magnetization and ϕ the angle between the applied magnetic field and the sample plane, which equals 45° in these experiments, as indicated on fig. 8.2.

$$\boldsymbol{\Omega} = \frac{g^* \mu_B}{\hbar} (\mathbf{B} + \mathbf{B}_N) \quad (8.3)$$

with g^* the effective g-factor, μ_B the Bohr magneton and $\hbar$ Planck's constant and $\mathbf{B}$ the external field.

The total field that the electron spins experience is the superposition of the external field $\mathbf{B}$ and the nuclear field $\mathbf{B}_N$, which can be described as follows:[1]

$$\mathbf{B}_N = b_N \cdot \mathbf{B} \cdot \frac{\mathbf{s} \cdot \mathbf{B}}{B^2 + B_{loc}^2} \quad (8.4)$$

with $\mathbf{s}$ the unit vector in the direction of the average spin, $\mathbf{B}_N$ the effective nuclear field acting on the electron spins, B_{loc} the local random field that

the nuclei experience and b_n is the (scalar) high-field value of $\mathbf{B}_N$ (when $\mathbf{s} \parallel \mathbf{B}$ and $B \gg B_{loc}$):

$$b_N = f \cdot b_{ni} \frac{I(I+1)}{S(S+1)} \langle S \rangle \quad (8.5)$$

with $S = 1/2$, $I = 3/2$. $f = \frac{T_1}{T_1 + T_{1e}}$ is the leakage factor where T_{1e} and T_1 are respectively the nuclear relaxation time due to the hyperfine interaction with the spin-polarized electrons and the nuclear relaxation time due to other mechanisms. $\langle S \rangle$ is the average electron spin. For one isotope b_{ni} can be expressed as follows:[1]

$$b_{ni} = \frac{16\pi}{3g^*v_0} \mu_I \eta \xi \quad (8.6)$$

with μ_I the nuclear magnetic moment, v_0 the volume of one unit cell, η the electron density in one unit cell and ξ the number of nuclei of the considered isotope in the unit cell. The measured nuclear field is always the sum of the respective nuclear fields of the different isotopes, so we use a single b_{ni} .

8.2.2 Transient effects

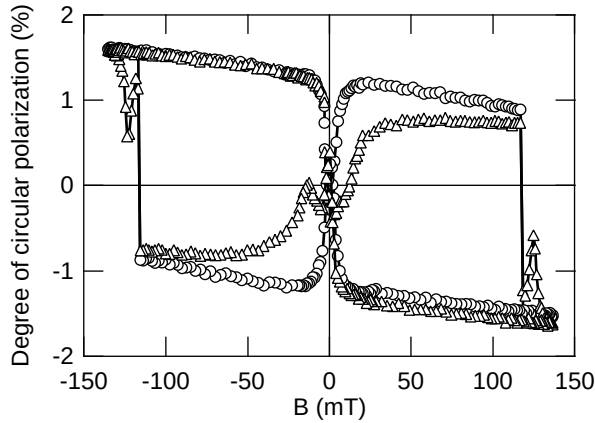


Figure 8.3: The circular polarization of the electroluminescence as a function of the external field at 5 K and 750 A/cm². ($\triangle$): fast measurement: 1 datapoint per second, ($\circ$): slow measurement: a pause of 60 s after changing the external field.

Initial observations

Fig. 8.3 shows the measured degree of circular polarization of the electroluminescence of the sample as a function of the oblique magnetic field. The circles are measurement data from a “slow” scan: after adjusting the value of the magnetic field, there is a pause of 1 minute before measuring the degree of circular polarization, while the current is continuously flowing through the device. The triangles represent a “fast” scan: in this case there is a pause of only 1 second after a change of the field. The slow measurement shows a Lorentzian shaped curve, which is characteristic for an oblique Hanle measurement. The saturation field of the circular polarization is only about ~ 10 mT, which, without the assumption of a nuclear field would imply a very long electron spin lifetime (~ 10 ns). An independent optical pumping experiment (not shown)[9], however, showed a much smaller $T_S \sim 200$ ps. The reason for this discrepancy is the presence of a strong nuclear field that results in an external field dependence mimicking the oblique Hanle effect signature: it accelerates spin precession and saturates the optical polarization already in small fields, but vanishes around zero field. This is furthermore corroborated by the findings of the fast measurement. This measurement looks quite different, there are transient effects present at the reversal of the magnetic field and at the reversal of the MnSb magnetization. It seems that at these points, the effective magnetic field caused by nuclear spins changes more slowly than the applied magnetic field. To clarify these effects, we have performed three types of measurements.

- 1. The sign of the injected electron spins is changed, keeping the external magnetic field constant.
- 2. The sign of the injected electron spins is kept constant, but the external magnetic field is reversed.
- 3. Both the injected electron spins and the external magnetic field are reversed.

During the changes we switch off the device current, such that during the reversing of the magnetic field, or the reversing of the magnetization, the electron spins don’t influence the nuclear spins. Immediately after the manipulation, the current is switched back on and the circular polarization of the light is monitored as a function of the laboratory time.

We will now discuss the three experiments. In a first measurement, shown in Fig. 8.4, the MnSb film is saturated in a large positive field, after which the magnetic field is set to -100 mT. Next, after switching off the current through the device, the MnSb magnetization is reversed by sweeping the field to -130 mT and returning to -100 mT. After this, the device

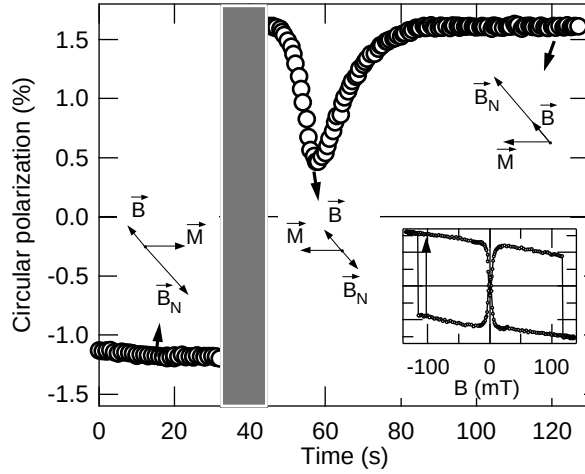


Figure 8.4: The degree of circular polarization of the electroluminescence vs time after reversing the magnetization of the MnSb in a negative external field at 5 K and 750 A/cm^2 . During the gray period the current through the device is switched off and the magnetization is reversed. A magnetization reversal in a positive external magnetic field gives the same result. The bottom right inset shows the full Hanle curve, which schematically shows that we reverse the MnSb-magnetization. The vector diagrams show the relative orientation of the magnetization $\vec{M}$, the applied field $\vec{B}$ and the nuclear field $\vec{B}_N$ during the time sweep.

current is switched back on and the circular polarization of the light is monitored. The data show that the circular polarization instantaneously reverses, reflecting the change in sign of the injected spin polarization, then slowly decreases, reaches a minimum and increases again up to its original value. This measurement indicates that the nuclear spins, which haven't changed during the dark period, are slowly reversed by the electron spins. This causes the nuclear field and thus the total field acting back on the electron spins to decrease, reverse sign and increase again, resulting in a dip in the circular polarization.

In Fig. 8.5 a similar experiment is shown, but instead of the electron spins, the magnetic field is reversed in the dark period. The field is swept from -63 mT to $+63 \text{ mT}$. The field sweep ($\sim 4 \text{ mT/s}$) is slow in comparison to the transverse nuclear spin lifetime T_2 ($\sim 100 \mu\text{s}$), which means that at all times, the nuclear spin ensemble is in internal equilibrium. A characteristic

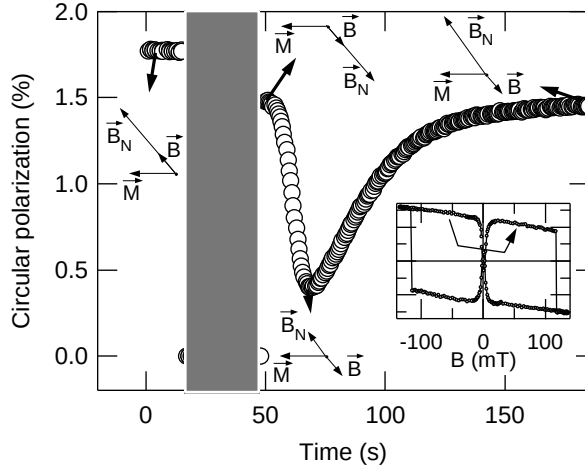


Figure 8.5: The degree of circular polarization vs time after reversing the external field from -63 mT to +63 mT at 5 K and 750 A/cm². During the gray period the current through the device is switched off and the field is reversed. Reversing the field from +63 mT to -60 mT gives the same result. The bottom right inset shows the full Hanle curve, which schematically shows that we reverse the sign of the external magnetic field. The vector diagrams show the relative orientation of the magnetization $\vec{M}$, the applied field $\vec{B}$ and the nuclear field $\vec{B}_N$ during the time sweep.

dip similar to the previous experiment appears in the circular polarization. The appearance of this dip suggests that during the reversal of the magnetic field, the nuclear spins have changed sign. After the electron spin injection is switched on again, the electron spins can slowly force the nuclear spins back to their original position, i.e. aligned with the electrons spins. To check this hypothesis, we have performed a similar measurement, but in this case reversing both the electron spins and the magnetic field.

Figure 8.6 shows that when the field is swept from -30 mT to +130 mT, no transient effects occur in the degree of circular polarization. Because both the electron spins and the nuclear spins have reversed in the dark period, no transient behavior is expected.

Measurements of the nuclear relaxation time and the nuclear field

The slow reversal of the nuclear field due to the hyperfine interaction allows us to measure the nuclear spin relaxation time due to the hyperfine interac-

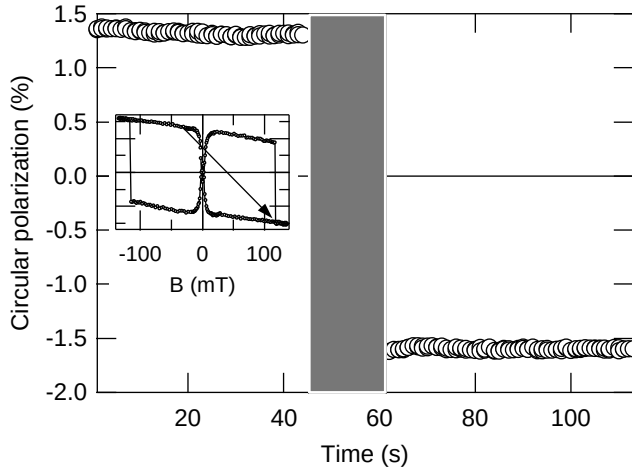


Figure 8.6: The degree of circular polarization of the electroluminescence vs time after reversing both the external field and the MnSb-magnetization at 5 K and 750 A/cm². The inset shows the full Hanle curve, which schematically shows that we reverse the sign of both the external magnetic field and the MnSb magnetization.

tion. The rate equation for the evolution of the nuclear spin system is given by:[10]

$$\frac{d\langle I_z \rangle}{dt} = -\frac{1}{T_{1e}} \left[\langle I_z \rangle - \langle S \rangle \cdot \frac{I(I+1)}{S(S+1)} \right] - \frac{1}{T_1} \langle I_z \rangle \quad (8.7)$$

with $\langle I_z \rangle$ the average nuclear spin parallel to the magnetic field and $\langle S \rangle$ the average electron spin. This leads to a steady-state average nuclear spin given by:

$$\langle I_z \rangle = \frac{I(I+1)}{S(S+1)} \cdot \frac{T_1}{T_{1e} + T_1} \cdot \langle S \rangle \quad (8.8)$$

Giving that the GaAs active layer is not intentionally doped and that the electroluminescence spectrum shows band-to-band emission, the dominant nuclear relaxation mechanism is due to free (conduction) electrons. This implies that the relaxation time T_{1e} is given by:[11]

$$T_{1e}^{-1} = \frac{32}{9} \gamma_e^2 \cdot \gamma_n^2 \cdot n \cdot \eta^2 (2\pi m^3 kT)^{\frac{1}{2}} \quad (8.9)$$

With γ_e and γ_n the electronic and the nuclear gyromagnetic ratios, respectively. n is the carrier concentration and η the electronic density at the

nucleus.

Using GaAs parameters[1], at a temperature of 5 K, and a carrier concentration of $4 \cdot 10^{17} \text{cm}^{-3}$, we get an estimated $T_{1e} = 27$ s.

When the injected electron spins have changed sign due to the reversing of the MnSb magnetization or when the nuclear polarization has been adiabatically reversed, the nuclear polarization (corresponding originally to a nuclear field b_{N0}) will, according to eq. 8.7, exponentially evolve towards its new equilibrium value (corresponding to a nuclear field b_{N1}), with a time constant T_{1e} . To fit the curves on figure 8.4 and 8.5, we use eq. (8.2), with a nuclear field that varies exponentially with time using a time constant τ that combines both T_{1e} and T_1 .

$$b_N = b_{N1} - (b_{N1} - b_{N0}) \cdot \exp(-(t - t_0)/\tau) \quad (8.10)$$

We have used the nuclear field at the start of the sweep b_{N0} , at the end of the sweep b_{N1} and the time constant τ as fit parameters. The injected spin polarization is determined from the full Hanle curve and the spin lifetime T_S in the GaAs active region is determined to be 200 ps by an independent optical pumping experiment (not shown).

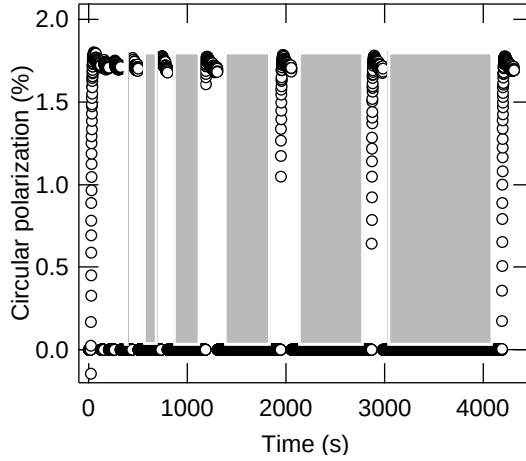


Figure 8.7: Nuclear depolarization curve at 5 K and 18 mT. The device current is switched off during increasing time intervals (grey areas) and switched on again.

Nuclear spin-lattice relaxation time T_1 To investigate the decay time of the nuclear polarization due to direct coupling to the lattice, without the

presence of electrons, we monitored the circular polarization of the light, while we switched off the current during increasing time intervals. By analyzing the initial circular polarization after switching the current back on (Fig. 8.7), the nuclear spin-lattice relaxation time T_1 can be deduced. During the time that the current is switched off, T_{1e} can be considered as infinite and τ equals T_1 . From a measurement shown on fig. 8.7 at 4.6 K and 17.8 mT, T_1 can be deduced to be near 7.5 min. This time strongly depends on the value of the external magnetic field, as shown in figure 8.8.

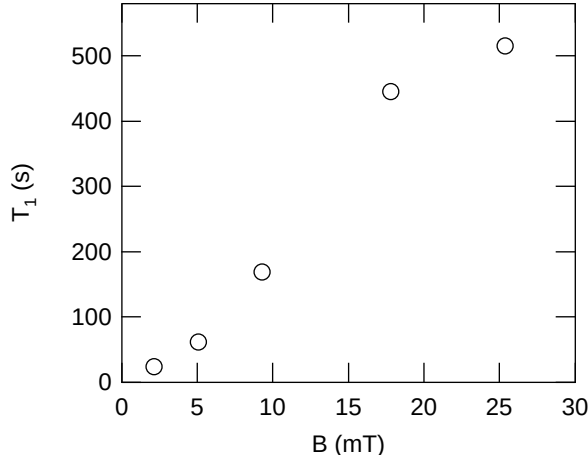


Figure 8.8: The nuclear spin-lattice relaxation time T_1 as a function of the external magnetic field at 5 K.

The nuclear spin relaxation time due to the hyperfine interaction
 T_{1e} To investigate the hyperfine interaction time T_{1e} , we analyze the transient effects that occur in figures 8.4 and 8.5. In order to obtain a measure of T_{1e} , we use a measurement at relatively high field, where $T_1 \gg T_{1e}$, such that τ equals T_{1e} . It appears that there is a non-homogeneous distribution of the time constant over the sample. If the switching of the nuclear polarization could be described by just one time constant and a homogeneous nuclear field, at one point during the nuclear spin reversal process the nuclear field would exactly cancel the external field and the circular polarization should go through zero. We haven't observed such a phenomenon, which indicates that the nuclear polarization is not homogeneous. We have used a linear distribution for the time constant to take this into account. The fit of the transient curve on figure 8.9 gives a time constant that is

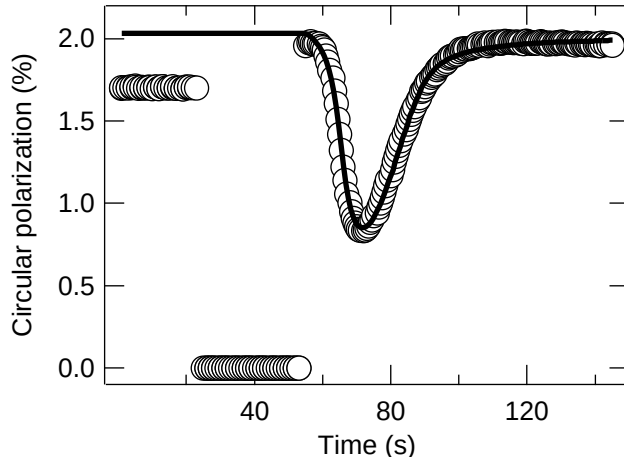


Figure 8.9: Transient behavior of the circular polarization after reversing the external magnetic field at 5 K and 750 A/cm^2 ($\circ$), together with the fit (solid line) based on eq. 8.10. Fit parameters: $T_{1e} = 24 \text{ s}$, $b_N \sim 1 \text{ T}$.

distributed between 24 s and 60 s. The weight is maximal at 24 s and decreases linearly towards zero at 60 s. The fit value of the nuclear field is about 1 T. The inhomogeneous distribution of the nuclear polarization may be explained by a non-homogeneous distribution of the carrier concentration in the active GaAs region. In fig. 8.10 the simulated electron concentration is shown in the active region as function of the depth. The simulations show that the electron concentration is not uniform across the active region but has a maximum near the top AlGaAs barrier.

Temperature and bias dependence of the time constant T_{1e} The characteristic time constant of the hyperfine interaction T_{1e} decreases with temperature, as shown on figure 8.11 and indicated by eq. 8.9. The behavior as expected from eq. 8.9 is shown by the solid line. There are a few reasons why the actual behavior deviates from the expected. There is an error on the temperature read-out. Since the current density through the device is quite high, the actual temperature in the device is probably several degrees above the measured value. Furthermore, the expected behavior is based on a carrier concentration of $4 \cdot 10^{17} \text{ cm}^{-3}$. As is shown on figure 8.10, the electron concentration within the active region is very inhomogeneous. This causes a inhomogeneous nuclear polarization and time constant within the active region. It is difficult to estimate how the effective electron concentration

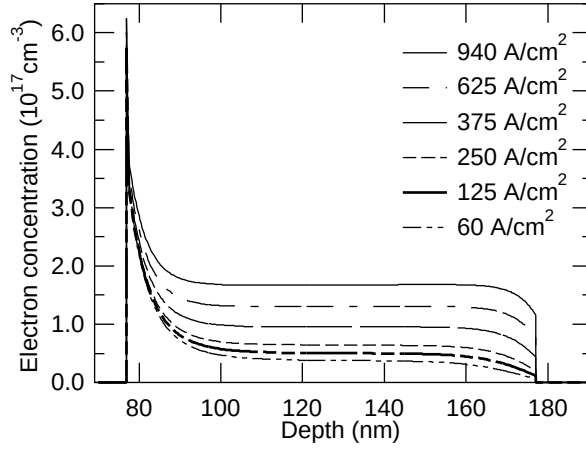


Figure 8.10: The simulated electron concentration in the GaAs active region as function of the depth for different values of the current density. The concentration is the highest near the top AlGaAs barrier.

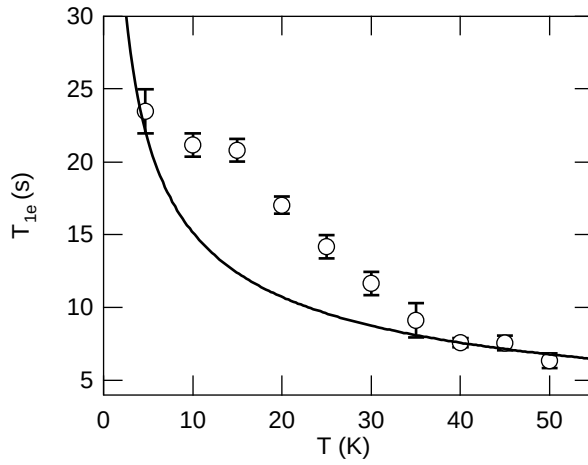


Figure 8.11: T_{1e} as function of the temperature ($\circ$) at 750 A/cm^2 , together with the theoretical expected behavior (solid line) based on eq. 8.9.

changes as function of the temperature.

According to eq. 8.9 the time constant $T_{1e} \sim n^{-1}$. Higher injection

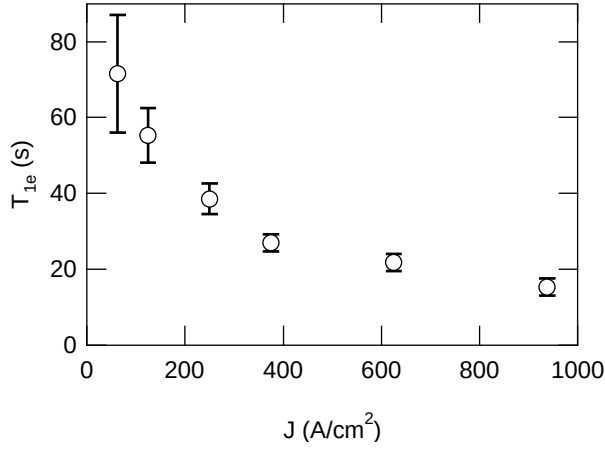


Figure 8.12: T_{1e} as a function of the bias current density at 5 K.

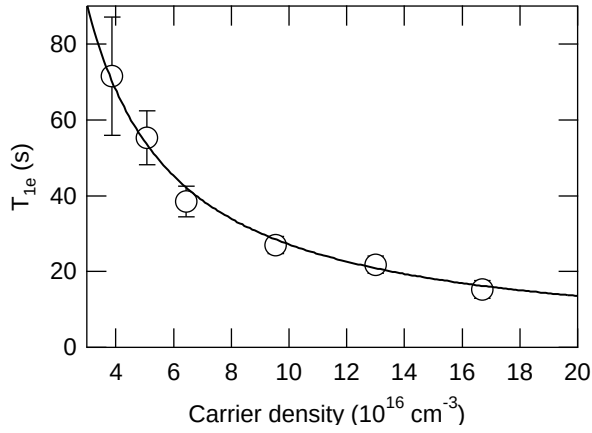


Figure 8.13: T_{1e} as a function of the simulated electron density ($\circ$) and the theoretically expected behaviour based on eq. 8.9 at 5 K (solid line).

currents hence lead to smaller T_{1e} . On figure 8.12 the experimental T_{1e} is shown as a function of the applied current. Indeed T_{1e} decreases with increasing bias, consistent with eq. 8.9. The plotted T_{1e} corresponds to the time constant with the maximal weight in the fit. At lower bias the distribution of the time constants shifts towards larger time constant, as indicated

in figure 8.12, but the distribution still has a tail at lower time constants (to about 10 s). This could be explained by the non-homogeneous distribution of the electron concentration in the active GaAs region. Although the average concentration in the active region changes strongly with applied bias, the peak concentration near the top AlGaAs barrier is less sensitive to the bias, as is shown on fig. 8.10. Using the electron density given by the simulations on Fig. 8.10, we have plotted T_{1e} versus the electron density on Fig. 8.13. It is clear that T_{1e} is inversely proportional to the electron density in the active region, as implied by eq. 8.9.

Bias dependence of the nuclear field b_N The fitting procedure of the transient effects of the electroluminescence allows not only to obtain a value of the time constants involved, but also the absolute value of the nuclear field. In figure 8.14 the dependence of this fitted nuclear field is shown as a function of the bias. At low values of the nuclear field we can verify this procedure by direct visualization of the nuclear field. When the magnetization and the external magnetic field are aligned anti-parallel the nuclear field is aligned anti-parallel to the external field. The effective field for the electrons is the superposition of both the external magnetic as the nuclear field. When the external field equals the nuclear field, there is no precession and hence there should be no circular polarization. Due to the limited coercivity of the MnSb-film, this field cannot be reached. However, due to the slow response of the nuclear spin system upon external changes, we can bypass this problem. When we use the procedure described in section 8.2.2 and figure 8.4 to visualize the transient effects after a reversal of the magnetization, and sweep the magnetic field to different values before switching on the device current, we can monitor the initial behavior of the circular polarization as function of the time for different values of the magnetic field. If the external field is smaller than the nuclear field when the device current is switched back on, the current induced reversal of the nuclear field causes a dip in the circular polarization, since the effective field for the electrons first decreases until $B = B_N$, before increasing again. If the external field exceeds the nuclear field, only an increase of the polarization is expected, since the effective field for the electrons only increases in this case. This is shown on figure 8.15 and figure 8.16 for different values of the current density. On figure 8.15 there is a transition from a clear dip in the circular polarization to just a slight increase of the circular polarization, while in the case of figure 8.16 this transition is just above the highest applied field. When we then put these values in figure 8.14, they confirm the values obtained by the fitting procedure.

The bias dependence of the nuclear field can be understood based on eq. 8.8. The average nuclear spin is proportional to both the average electron

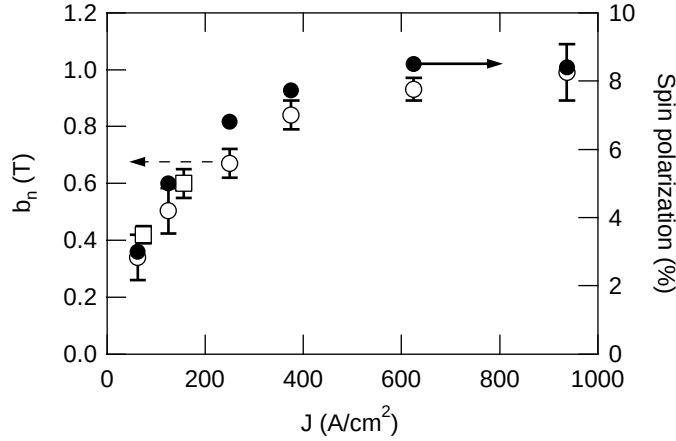


Figure 8.14: b_N as a function of the bias current density at 5 K obtained through the fitting procedure (o) and through the field-dependence of the transient effects (□) and the steady state electron spin polarization as function of the applied current density at 5 K (•).

spin $\langle S \rangle$ and the factor $\frac{T_1}{T_1 + T_{1e}}$. Since the measured nuclear field has been determined at a relatively high external field, where T_1 exceeds 500 s, the average nuclear spin is determined by the average electron spin. This is confirmed by a measurement of the bias dependence of the steady-state electron spin polarization in the GaAs active region, as shown by the bullets (•) in figure 8.14. The steady-state electron spin polarization has been obtained by fitting the full Hanle curves using eq. 8.2. The increase of the nuclear field is clearly accompanied by an increase of the steady-state electron spin polarization.

8.2.3 Nuclear Magnetic Resonance

Unambiguous proof of the presence of a non-equilibrium nuclear polarization can be given by investigating the effect of an oscillating magnetic field that brings the nuclei into resonance, Nuclear Magnetic Resonance (NMR).[12; 13] We inserted a coil in the cryostat to generate an ac-field which is perpendicular to the direction of the injected spins and to the applied dc-field. The ac-signal is generated by a signal generator and amplified by a broad-band amplifier.

In order to completely destroy the nuclear polarization the frequency was swept continuously through the resonance frequencies of the different

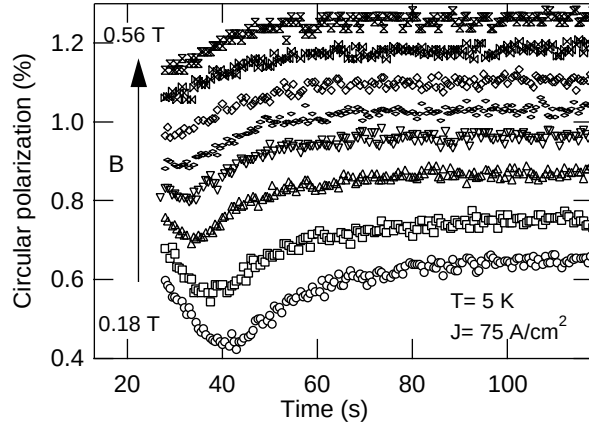


Figure 8.15: Transient effects in the circular polarization upon a reversal of the magnetization for different values of the external field at a current density of 75 A/cm^2 and a temperature of 5 K . The applied field goes from 0.18 T to 0.56 T . The curves are offset for clarity.

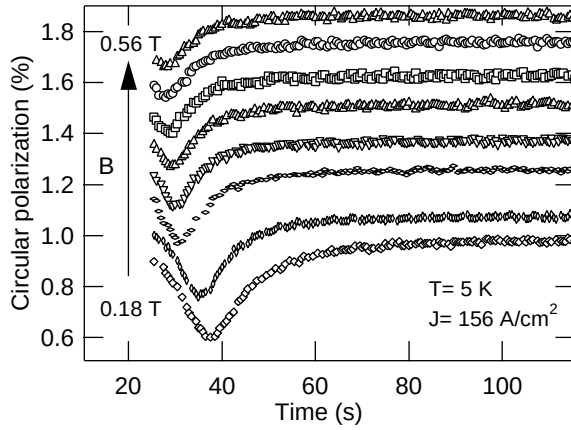


Figure 8.16: Transient effects in the circular polarization upon a reversal of the magnetization for different values of the external field at a current density of 156 A/cm^2 and a temperature of 5 K . The applied field goes from 0.18 T to 0.56 T . The curves are offset for clarity.

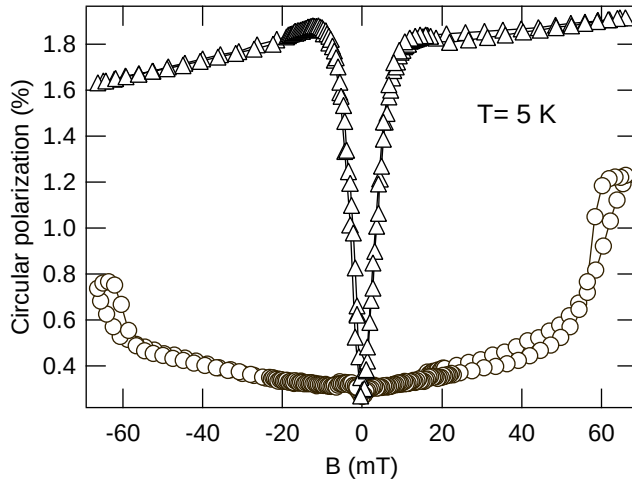


Figure 8.17: The Hanle curve without the application of an ac-field (Δ) and while sweeping once per second from 40 to 700 KHz ($\circ$).

isotopes (^{69}Ga , ^{71}Ga and ^{75}As) at 5 K. Figure 8.17 shows the oblique Hanle measurement without applied ac-field ($\circ$) and while sweeping the frequency between 40 and 700 KHz once a second (Δ). At low dc-fields, when the resonance frequencies were within the scanning range, the measured circular polarization was completely quenched, indicating a much smaller effective field acting on the electron spins.

Keeping the external field constant and sweeping the frequency slowly through the resonance frequencies allows to visualize the different isotopes. Figure 8.18 shows the circular polarization as a function of the frequency of the ac-field at 80 K and 35 mT. The As and Ga isotopes can be clearly distinguished and appear at the expected frequency. This is further confirmed when the resonance frequencies are plot as a function of the applied field on figure 8.19. The resonance frequencies show a linear behavior with magnetic field: 1270 Hz/Oe for ^{71}Ga , 1006 Hz/Oe for ^{69}Ga and 708 Hz/Oe for ^{75}As , in good agreement with the gyromagnetic ratios: $\gamma(^{71}\text{Ga}) = 1298 \text{ Hz/Oe}$, $\gamma(^{69}\text{Ga}) = 1022 \text{ Hz/Oe}$ and $\gamma(^{75}\text{As}) = 729 \text{ Hz/Oe}$. [1]

At 5 K, the situation changes, the ^{71}Ga -nuclei dominate the nuclear field and the ^{75}As -resonance can not even be observed. Figure 8.20 shows the measured circular polarization at 5 K and at an external field of 15 mT. The frequency was swept from low to high frequency in this measurement. Only the Ga-isotopes are clearly visible on this scan. The ^{71}Ga -signal is the largest one, while the ^{69}Ga is substantially smaller. The ^{75}As -signal

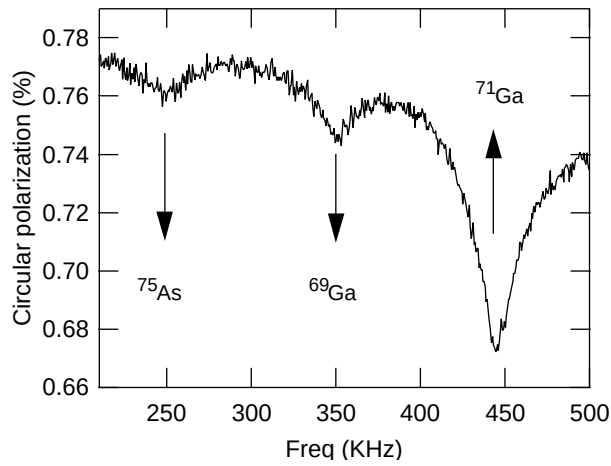


Figure 8.18: The circular polarization of the electroluminescence as a function of the frequency of the ac-field at 80 K and 35 mT. At the resonance frequencies of the GaAs-isotopes a dip in the circular polarization is clearly visible.

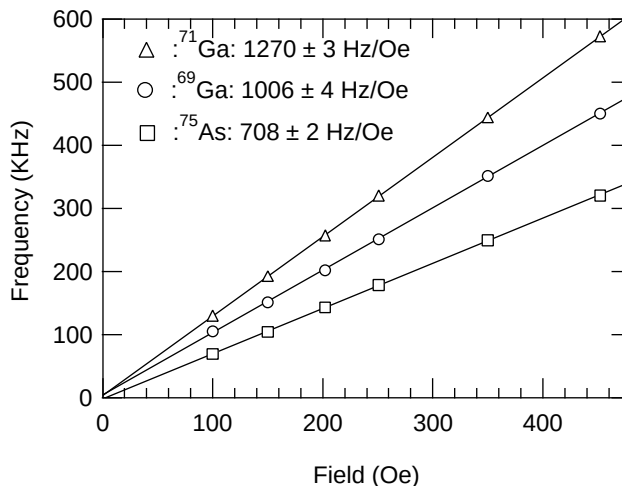


Figure 8.19: The measured resonance frequencies of the three isotopes in GaAs as a function of the applied field at 80 K.

is very weak and can be hardly distinguished. This has been consistently observed for different scan speeds, scan directions and values of the external field. The absence of the ^{75}As -signal means that the induced electron spin precession due to the sum of the ^{71}Ga and the ^{69}Ga -field is fast enough to saturate the circular polarization. Small changes to the total field because of a change of the ^{75}As -field hence have little influence on the degree of circular polarization. This indicates that mainly the ^{71}Ga -nuclei and to a smaller extent the ^{69}Ga -nuclei are responsible for the nuclear field.

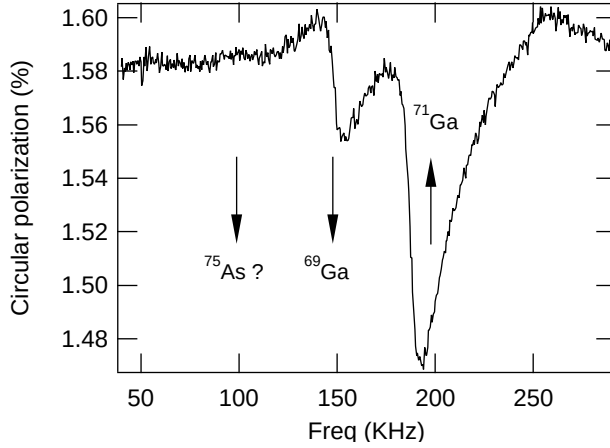


Figure 8.20: The circular polarization of the electroluminescence as a function of the frequency of the ac-field at 5 K and 15 mT while sweeping from high to low frequency. The resonance signatures of ^{71}Ga and ^{69}Ga can be distinguished.

8.3 Discussion

8.3.1 The field induced nuclear reversal

The nuclear field reversal during the magnetic field reversal (Fig. 8.5 and 8.6) appears to be faster than the time resolution of the measurement and indicates an adiabatic process. The field is swept sufficiently slowly such that the nuclear spins can at all times follow the field. Take for example a nucleus that is initially aligned (anti-aligned) with the field. When the applied magnetic field is lowered, the total field seen by the nucleus is the vector sum of $\mathbf{B}$ and a local field $\mathbf{B}_{loc}$. This local field consists of the random dipolar fields induced by the neighboring nuclei *and* the stray field

of the MnSb ferromagnetic layer. A magnetostatic simulation shows that this stray field is of the order of 0.2 mT, which is of the same order as the dipolar field. Since we switch off the electron injection during the zero-field traversal, the nuclei don't feel the electronic field. The magnetic field $\mathbf{B}$ is changed slowly enough compared to the Larmor precession cycle such that the nuclei will remain aligned (anti-aligned) with the total field. At zero external field, the nucleus will be pointing parallel to $\mathbf{B}_{loc}$. The average nuclear spin of the sample at that point is zero, since the local fields point in random directions in different parts of the sample, but the degree of alignment of the individual spins with their local field is preserved.[14] If we now turn the magnetic field back on, but in the reversed direction, the nucleus will continue to be aligned (anti-aligned) with the total field, which causes a reversal of the nuclear spin polarization.

8.3.2 The dominance of ^{71}Ga and the low-field nuclear depolarization

At first point it is difficult to understand why the nuclear field is dominated by the ^{71}Ga -nuclei. Previous experiments and theoretical calculations showed that the ^{71}Ga -nuclei even exert the smallest effective field on the electrons ($b(^{75}\text{As}) = -18.4$ kG, $b(^{69}\text{Ga}) = -9.1$ kG and $b(^{71}\text{Ga}) = -7.8$ kG).[15] This leads to the conclusion that the ^{75}As -nuclei are only weakly polarized. A possible explanation lies in the different nuclear relaxation times of the different isotopes. It is known[16] that the most important nuclear relaxation mechanism in GaAs is the quadrupolar relaxation mechanism. The quadrupole moment of As is higher than that of Ga, which causes a smaller nuclear spin lifetime ($T_1(^{71}\text{Ga}) = 6.6 \times T_1(^{75}\text{As})$ and $T_1(^{71}\text{Ga}) = 2.3 \times T_1(^{69}\text{Ga})$). Since the steady-state nuclear polarization is an equilibrium between pumping and relaxation, the nuclear polarization of the As-spins can be significantly smaller when the pumping time is similar to the relaxation time.

More evidence for this is given by the field-dependence of the nuclear spin relaxation time at 5 K, which is shown on figure 8.8. The dark nuclear relaxation time strongly decreases with decreasing external field. At high field the nuclei are decoupled and the observed spin relaxation time is mainly given by the ^{71}Ga -spin relaxation time, while at lower fields the interaction between the Ga and the As spins increases such that there is an out-diffusion of nuclear spin polarization from the Ga spin bath to the ^{75}As spin bath. The spin relaxation time will then be dominated by the (short) ^{75}As -relaxation time. This can also explain why in previous reports[1; 15] the ^{75}As -resonance was much stronger. In their case the nuclear polarization was mainly created by electrons that were localized on donor sites. The characteristic hyperfine interaction time for localized electrons is a lot

smaller than for conduction electrons (~ 0.1 s). Hence, the shorter pumping time (i.e. an increased pumping efficiency) allowed a larger ^{75}As -steady state polarization.

The resonance signal of ^{75}As is nevertheless visible at 80 K. This could be explained by the overall smaller nuclear field. The nuclear field is not strong enough to completely saturate the Hanle curve such that small changes to the total nuclear field are more easily visible in a frequency scan.

8.3.3 The zero-field dip

The half-width of the zero-field dip in the Hanle-curve on figure 8.3 should correspond to the random local field that the nuclei experience (B_{loc} from eq. 8.4). Its value, measured in steady-state conditions, is about 7 mT at 4.5 K and 750 A/cm². This value is over 10 times higher than the optical orientation value of the local dipole field, which is more in the order of 0.2 mT.[1] Other reports also indicate that in different structures and different measurement geometries the local fields seem higher than measured with PL in bulk GaAs.[2; 13; 17; 18] A solution is offered by the field-dependence of the nuclear relaxation time as shown on figure 8.8. At low fields, the nuclear relaxation time becomes comparable to the pumping time induced by the hyperfine interaction with the electron spins. The leakage factor $f = \frac{T_1}{T_1 + T_{1e}}$ hence decreases strongly with decreasing field and causes a strong decrease of the steady-state nuclear spin polarization with decreasing field:

$$B_{N,lowfield} = \frac{T_1}{T_{1e} + T_1} B_{N,highfield} \quad (8.11)$$

under the assumption that $f \approx 1$ at high field.

To test this hypothesis we looked first at the bias-dependence of the half-width of the zero-field dip. In figure 8.21 the apparent local field B_{loc} is shown as a function of the current density. It's clear that the apparent B_{loc} decreases strongly with increasing electron pumping. B_{loc} has been obtained by fitting the full Hanle curves using eq. 8.2, combined with eq. 8.4. A similar behavior has been observed before at 80 K.[4] When the bias increases, the electron concentration increases and according to eq. 8.9 T_{1e} drops. The drop in T_{1e} hence narrows the dip in the Hanle curve.

In an effort to reconstruct the Hanle-curve without the presence of this low-field nuclear relaxation, we measure the circular polarization as function of time and start with the field high enough such that $T_1 \gg T_{1e}$. We subsequently quickly move the field to a low-field value and monitor the decay of the circular polarization as function of time. The value of the circular polarization immediately after the field change corresponds to the value "without nuclear relaxation". On figure 8.22 such an experiment is

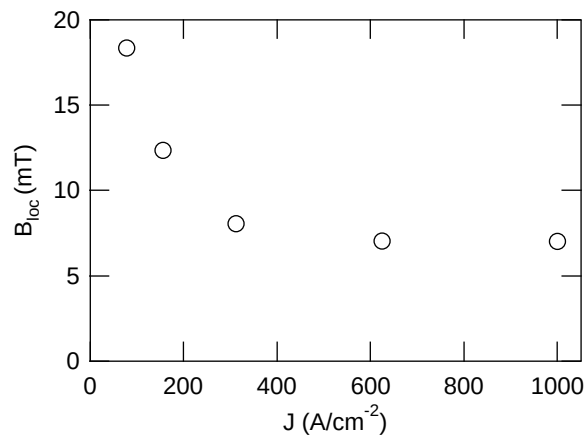


Figure 8.21: The apparent local field B_{loc} as a function of the current density at 5 K.

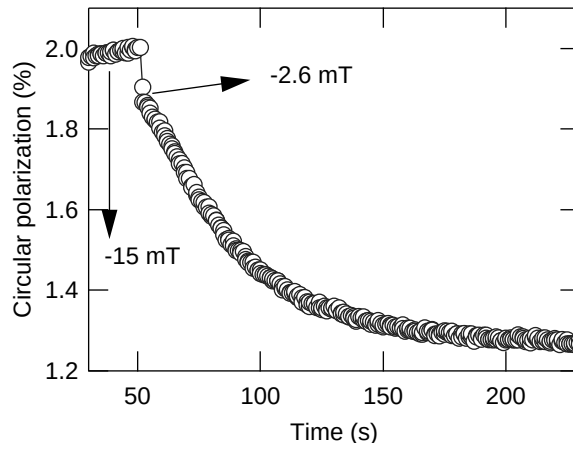


Figure 8.22: Transient measurement to visualize the enhanced low-field nuclear relaxation at 5 K. The initial value of the external field is -15 mT, which is then rapidly swept to -2.6 mT. From this time the circular polarization of the electroluminescence starts to decrease to eventually reach its steady-state value.

shown, where the field was -15 mT originally and has then been swept

rapidly to -2.6 mT. It's clear that the circular polarization slowly decays to its steady-state value. This procedure can be performed for many field-points around zero field and the result is a Hanle-curve with a narrow dip ($\Delta B \sim 0.5$ mT), as shown on figure 8.23.

The magnitude of the apparent local field in the steady-state measurements can hence be ascribed to the field dependence of the nuclear spin relaxation time. This causes a decrease of the nuclear field at low values of the external magnetic field and hence an increased apparent local field.

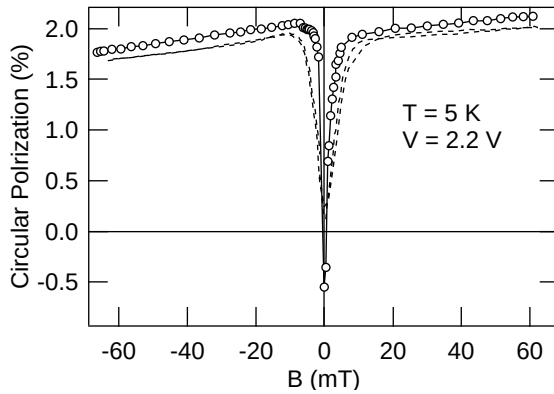


Figure 8.23: The reconstructed Hanle curve at 5 K obtained using the method demonstrated in figure 8.22 ($\circ$) and the steady state Hanle curve (dashed line).

Gammon *et al*[18] also measured an enhanced nuclear relaxation at relatively large values of the external field in GaAs quantum dots. They attributed this effect to an effective magnification of the local field, mediated by fluctuating hyperfine fields of electron spins. In our experiment, however, the enhanced nuclear relaxation is measured by switching off the current through the device and monitor the initial polarization after switching the device current back on. In this case, while the device current is switched off, the active region is located within the depletion region of the $p-n$ -diode and is depleted of carriers. Hence it is hard to imagine that the electron spins mediate the enhanced nuclear relaxation through the hyperfine interaction. Nevertheless, it appears that the zero-field dip strongly depends on the exact experimental configuration and sample geometry.

8.3.4 The local field

The dip in figure 8.23 is asymmetric around zero field and the circular polarization becomes briefly negative during the field sweep. This last aspect can only be explained assuming an additional field in the semiconductor. First of all, as already discussed, the local field seen by the nuclei is given by the dipole field, the electronic field *and* the stray field from the ferromagnetic top layer. The dipole fields measured in earlier optical orientation measurements typically were measured in p-type layers co-doped with silicon in order to achieve an efficient localization of the carriers. The localization assured that the nuclei were effectively polarized. In our case the nuclear spin relaxation time indicates that the dynamical nuclear polarization is caused by nuclear relaxation due to delocalized conduction band electrons. So unlike a mechanism of fast polarization by nuclei neighboring an impurity atom and a slower polarization of the bulk atoms by nuclear spin diffusion[19] the nuclei are homogeneously polarized by the conduction electrons. Hence they all experience the same electronic field. Increasing the carrier concentration can increase the value of this electronic field, that's given by:[20]

$$\Delta H = \frac{8}{3}\pi \langle |u_F(0)|^2 \rangle \Omega_v \cdot \mu_B \cdot \Delta n \quad (8.12)$$

with $\langle |u_F(0)|^2 \rangle$ the squared Bloch function amplitude at the nucleus averaged on the Fermi surface, Ω_v the volume of the unit cell containing one Ga and one As atom, μ_B the Bohr magneton and $\Delta n = n^\uparrow - n^\downarrow$, the difference between spin-up and spin-down electron concentrations. Taking $\langle |u_F(0)|^2 \rangle = 5.8 \cdot 10^{25} \text{ cm}^{-3}$ for the Ga-atoms, $\langle |u_F(0)|^2 \rangle = 9.8 \cdot 10^{25} \text{ cm}^{-3}$ for the As-atoms¹⁰ and $\Delta n = 4 \cdot 10^{16} \text{ cm}^{-3}$ (using $n = 5 \cdot 10^{17} \text{ cm}^{-3}$ and an electron spin polarization of 8 %), the electronic field acting on the Ga nuclei is about 0.8 mT and the electronic field acting on the As nuclei is 1.35 mT. Still higher carrier concentrations only increase this electronic field. Due to the low polarization of the As-nuclei, only the Ga-value matters here.

Figure 8.24(a) shows a simulation using a local field of 0.5 mT and an additional stray field of 0.8 mT. It is schematically shown why this would lead to a negative circular polarization at low field: The nuclear spins always follow the total field acting on the nuclei and this field is the vector sum of the external field and the stray field. At an external field that is considerably larger than the stray field, the nuclear spins always follow the external field. At low field however, the total field rotates towards the stray field. Since the average electron spin is the projection of the average injected spin on the total field acting on the electrons (= external field $\mathbf{B}$ + stray field $\mathbf{b}_s$ + nuclear field $\mathbf{B}_N$), the circular polarization can become negative. The analysis taking into account the electronic field is similar and leads to a

similar curve. The difference between the value of 0.8 mT and the predicted 0.2 mT can be caused by the simultaneous effect of the stray field and the electronic field. However, we cannot conclude from these measurements whether the additional local field mainly originates from the MnSb-induced stray field, the electronic field or a combination of both fields.

8.4 Conclusions

We have investigated the pumping of nuclear spins due to electrical spin injection from a MnSb top contact through a Schottky tunnel barrier in a (Al,Ga)As spin-LED using the oblique Hanle effect. Upon reversal of the MnSb magnetization (i.e. the injected electron spin polarization) or the external magnetic field, the measured degree of circular polarization exhibited slow transient effects that are correlated with slow changes in the nuclear spin system. The nuclear spin system has been shown to adiabatically reverse when reversing the external magnetic field. Nuclear magnetic resonance has shown that mainly the ^{71}Ga -isotope is responsible for the observed effects. Especially the ^{75}As -isotope is only weakly polarized, which we attribute to the more efficient quadrupolar spin relaxation of the As-nucleus.

Finally we have explained the enhanced width of the zero-field dip in the Hanle curve in our electrical pumping experiments as compared to optical pumping of compensated GaAs as the interplay between pumping and relaxation of the nuclear spins.

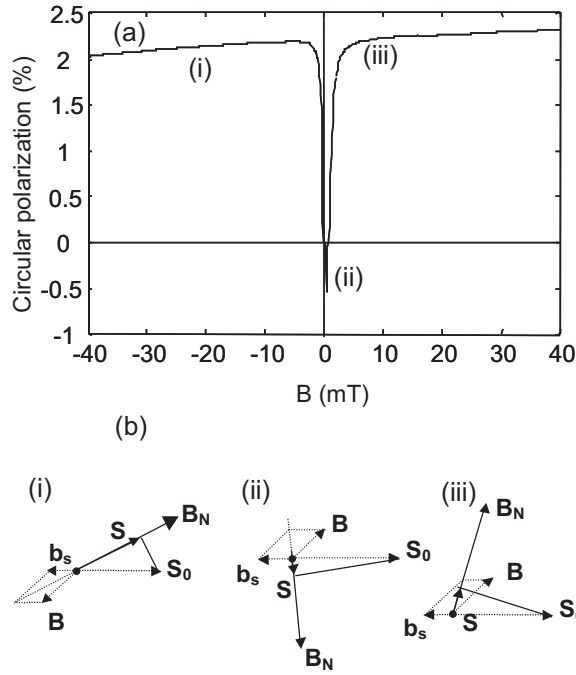


Figure 8.24: (a) Simulation of the Hanle curve at 5 K, taking into account a stray-field of 0.8 mT. (b) The different parts of the curve are schematically explained: (i) The average injected spin is in-plane, pointing to the right. The stray field $\mathbf{b}_s$ induced by the MnSb-film points to the left. The total field experienced by the nuclei is the sum of the external field $\mathbf{B}$ and the stray field $\mathbf{b}_s$ and is close to the external field. Hence, the direction of $\mathbf{B}_N$ is close to the direction of $\mathbf{B}$. The average electron spin $\mathbf{S}$ is the result of a projection of the injected electron spin $\mathbf{S}_0$ on the total field that acts on the electrons ($\sim \mathbf{B}_N$ when $\mathbf{B}_N$ is large). The observed circular polarization corresponds to the perpendicular component of the average electron spin and is positive in this case. (ii) At low (positive) values of $\mathbf{B}$, the total field that the nuclei experience and hence $\mathbf{B}_N$ points in the second quadrant. The perpendicular component of $\mathbf{S}$ then lies in the negative direction. (iii) $\mathbf{B}$ is again a lot stronger than the stray field and $\mathbf{B}$ lies again close to $\mathbf{B}$.

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Chapter 9

All-electrical spin injection and detection

9.1 Introduction

The previous chapters of this thesis all focused on electrical spin injection in GaAs from ferromagnetic materials and related phenomena. Electrical spin *detection* of spins in semiconductors by ferromagnetic materials can be regarded as the reverse process, although different in some points. Up till now, convincing results of electrical spin injection and detection in a lateral device are still lacking. In order to achieve this, a careful design of the ferromagnetic contacts, the semiconductor structure and a well-designed process flow is necessary.

In this chapter we focus on all-electrical spin-injection-detection devices. We first discuss previous experiments that have tried to show electrical detection of optically injected spins and concentrate afterwards on the design of an all-electrical device. We optimize the semiconductor doping profile, based on two-dimensional self-consistent simulations and develop a process flow to realize the devices.

9.2 Electrical spin detection of optically injected electron spins.

Electrical detection of spins in semiconductors has been pursued intensively in the last decade. As an analogue to the spin-light emitting diode, an electrical detection scheme can be set up by optically exciting a spin-polarized ensemble and detecting the spin density by a nearby ferromagnetic elec-

trode. Already in 1995 Prins and coworkers performed measurements on such a system[1]. They considered a Co/Al₂O₃/p-GaAs tunnel diode under optical excitation with modulated circularly polarized light and monitored the helicity dependent photocurrent as function of the incident photon energy, under the application of a perpendicular magnetic field of 0.4 T. This type of measurement is subject to large background signals. The modulation of the light not only creates a modulated density of spin-up and spin-down electrons in the semiconductor, but the σ^+ and the σ^- components of the light are absorbed differently in the ferromagnetic layer, generating an intensity modulation of the incident light with the same frequency as the polarization modulation. Hence, the helicity-dependent signal can easily be mistaken for a spin-dependent effect. The magneto-absorption and the spin-dependent effects are furthermore both proportional to the longitudinal component of the magnetization, making it difficult to distinguish between them. Although the spin-dependent effects were masked by magneto-optical effects, based on the excitation energy dependence of the helicity dependent photocurrent, they claimed to have measured a spin-dependent current modulation of 0.7%.

After this initial experiment, several follow-up experiments have been undertaken, such as using GaAs scanning tunneling microscope (STM) tips under the excitation with circularly polarized light, as spin-polarized sources to examine ferromagnetic materials at a local scale. Conclusive results, however, have not yet been achieved [2]. Also several experiments using optical excitation of GaAs in planar tunnel or Schottky contacts with ferromagnetic electrodes have been reported. Although some evidence for spin-dependent transport has been collected[3], the magneto-optical effects generally dominated the signal [4].

In a recent experiment, however, Crooker *et al* imaged the spin transport in a lateral two-terminal spin-injection device using the magneto-optical Kerr effect [5]. The distance between the two ferromagnetic electrodes exceeded the spin-diffusion length, excluding all-electrical spin injection and detection to occur. They were able to monitor the lateral decay of the injected spin polarization moving away from the injection contact *and* the build-up of a spin polarization moving towards the detection contact. The latter effect is caused by selective extraction of electron spins, leading to a spin accumulation at the semiconductor/metal interface. Furthermore, by optically exciting spin-polarized carriers with a low energy laser spot on a varying distance of the detection electrode, they were able to show conductance variations that can be ascribed unambiguously to spin-dependent transport. The advantage of this lateral excitation is the lack of the magneto-optical background effects, present in the previous reports. The observed signal, however, was very low and can probably be ascribed to the low amount of excess carriers created by the laser beam in the n-type

GaAs, generating only a small spin-polarization.

An important aspect of the electrical spin detection-experiments based on optical excitation of spin-polarized carriers in GaAs, is the escape-route for the minority electrons. By light illumination a constant amount of electron-hole pairs is created in the semiconductor. When the electrons flow to the metal/semiconductor interface, the spin-down electrons are reflected and accumulate near the interface. When there is no leakage path for these electrons to go to, they accumulate, flip and can enter the metal, resulting in a constant current, independent of the helicity of the light.[6] In p-type GaAs, e.g., this leakage path can be provided by recombination with holes, depending on the bias conditions of the device.

9.3 Spin transport in semiconductors, electric field dependence.

In contrast to spin-injection-detection experiments in metals, where electric fields are screened by the high carrier density, upon injection the spin-polarized carriers can be subject to quite high electric fields, certainly when they are injected in the space-charge region of a Schottky diode, for instance. The initial theoretical description of spin transport in semiconductors by Schmidt *et al*[7], Rashba[8] and Fert and Jaffrès[9] dealt with diffusive transport, neglecting electric field effects, and are hence only valid in situations with low electric fields, e.g. under low bias. Yu and Flatté discussed the effect of a homogeneous electric field on spin diffusion in non-degenerate semiconductors.[10] In their theoretical framework they excluded the treatment of space charge regions and high-injection effects. From their calculations could be inferred that the downstream spin diffusion length (i.e. along the field direction) is enlarged substantially by the application of an electric field. Vice versa the upstream spin diffusion length (anti-parallel to the field) is shortened with orders of magnitude. This picture has been generalized to degenerate systems by D'Amico, including many body effects caused by Coulomb electron-electron interactions.[11] This is advantageous for spin injection experiments, as the electric field effectively decouples the ferromagnetic metal and the semiconductor, enhancing the spin injection efficiency, but may be deleterious for spin detection. As mentioned in the case of optical excitation, for spin detection to occur, the reflected minority electron spins should find a leakage path to go to. In the case of an all-electrical detection device, they should diffuse back to the injection contact, which in the case of purely diffusive transport is possible. However, even in the case of small device dimensions compared to the nominal spin diffusion length, an applied electric field can cause a strong decrease of the upstream spin diffusion length, even below the device length, prohibiting a way back

for reflected electrons. This effect may destroy any magneto-resistance, but has yet to be demonstrated experimentally.

9.4 Simulations

In this section we focus on all-electrical spin-injection-detection devices and we show, based on one-dimensional self-consistent simulations, that in order to realize all electrical spin injection and detection, the design of the contact is of crucial importance and cannot simply be copied from the contacts used in the spin-LEDs. As already discussed in Chapter 7, in a metal/GaAs or a metal/oxide/GaAs contact the Fermi level pinning at the GaAs surface causes the surface Fermi level to be pinned about 0.8 eV beneath the conduction band of GaAs. In order to be able to inject electrons, this potential difference must be accommodated by the barrier. In a Schottky tunnel injector, the GaAs is heavily doped near the metal/GaAs-interface. This creates a narrow depletion region that takes up the difference between the surface and the bulk Fermi level and already allows tunneling at low bias between the metal and the GaAs conduction band. In the case of an oxide barrier, the depletion region is too wide to tunnel through and an external bias voltage is needed to supply the necessary voltage drop over the oxide barrier. In the case of bipolar devices such as spin-LEDs, the accumulated holes provide the positive charge required to generate the high electric field over the oxide, while in a unipolar lightly doped n-type device, the high electric field cannot be generated over the barrier. This limits the application possibilities for oxide barriers in unipolar devices and we propose the combination of an oxide and a heavily-doped Schottky barrier for an all-electrical device. We will discuss the optimal design of the doping level in a lateral spin-injection-detection device, based on 2D self-consistent simulations. The doping of the tunnel contacts is optimized in order to achieve an optimal tunnel barrier resistance and the doping of the spin-transport region is engineered to obtain an optimal current distribution.

Unipolar devices. In full-electrical spintronic devices, holes are unwanted due their detrimental effect on the electron recombination and the spin lifetime. In GaAs one rather prefers a moderate n-type doping close to 10^{16}cm^{-3} due to the long spin lifetimes and spin diffusion lengths.[12] Hence we consider a spin-injection structure using a metal/oxide injector on a lightly doped semiconductor. The simulated band diagram at an applied bias of 0.4 V and the simulated current-voltage (IV) characteristic are depicted in Fig. 9.1. The band diagram shows that the depletion region is about 100 nm wide, and that the voltage drop mainly takes place over the semiconductor. The IV-characteristics show a rectifying behavior, with a

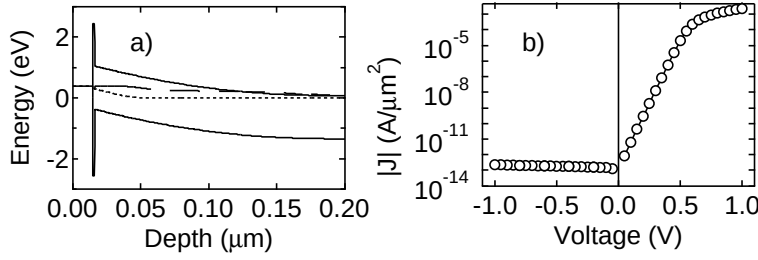


Figure 9.1: a) The band diagram at an applied bias of 0.4 V of a Metal/Oxide/GaAs tunnel diode with a lightly doped semiconductor: $N_D = 5 \times 10^{-16} \text{cm}^{-3}$. The dotted and the dashed lines represent the quasi-Fermi levels for electrons and holes, respectively. b) The simulated IV-characteristics of the device.

negligible current at reverse bias, indicating that the voltage drop over the oxide barrier is insufficient to allow tunneling in the simulated bias range. In contrast to the bipolar LED, no holes are available to accumulate at the oxide/GaAs interface. Hence the positive charge at the right side of the oxide barrier is supplied by a further depletion of the GaAs region farther away from the surface. This causes most of the voltage drop to occur in the GaAs region such that the voltage drop over the oxide is insufficient.

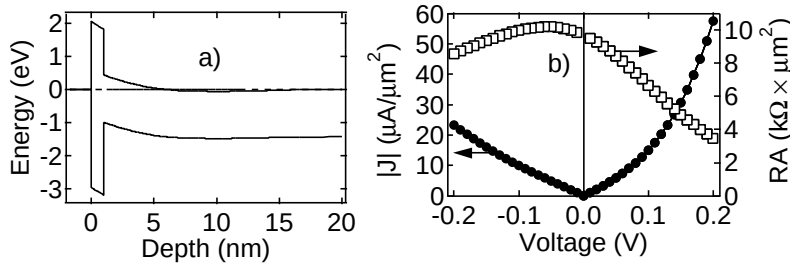


Figure 9.2: a) The band diagram at zero bias of a Metal/Oxide/GaAs tunnel diode with a heavily doped interfacial GaAs layer: 6 nm $N_D = 1.5 \times 10^{19} \text{cm}^{-3}$ /6 nm $N_D = 7 \times 10^{18} \text{cm}^{-3}$ and 1 nm AlO_x . b) The simulated current density ($\bullet$) and the resistance-area product ($\square$) of the device.

The use of a heavily doped sub-surface region allows to overcome this problem, as this narrows the depletion region and allows direct tunneling.

The use of an oxide barrier remains, however, preferential since it ensures the chemical stability of the ferromagnet/semiconductor interface and high spin-injection efficiencies have been shown for both AlO_x [13] and MgO -barriers.[14] Hence we propose the combination of an oxide barrier and a thin, heavily n-type doped semiconductor region that makes the Schottky barrier transparent and allows direct tunneling at low bias. We have chosen for a stepped doping profile: 6 nm $N_D = 1.5 \times 10^{19} \text{cm}^{-3}$ /6 nm $N_D = 7 \times 10^{18} \text{cm}^{-3}$ in order to ensure a narrow depletion region and to avoid a very high free carrier concentration in the non-depleted region. The band line-up is shown in Fig. 9.2(a). The depletion width is substantially smaller and the electrons tunnel directly into the semiconductor at low bias, and vice versa the electrons can tunnel into the ferromagnetic metal at a low value of the applied bias. This is shown in Fig. 9.2(b), where the current-voltage characteristics are symmetric around zero bias. The oxide thickness and the interfacial doping can be tuned in order to adjust the tunnel barrier resistance, and to reach an optimum magneto-resistance. According to Fert and Jaffrès[9], for the case of n-type GaAs, with a doping of 10^{16}cm^{-3} at room temperature, the optimal tunnel barrier resistance for a 1D-device is about $7 \text{ k}\Omega\mu\text{m}^2$. Fig. 9.2(b) shows that the tunnel barrier resistance is very close to this number, although slightly asymmetric with regard to the injection (reverse) and the detection (forward) polarities. This is intrinsic to the triangular Schottky tunnel barrier in the device, but can be ignored at small bias ($V < 0.1 \text{ V}$).

The tunnel resistance, though, depends exponentially on the tunnel barrier thickness, which is inversely proportional to $\sqrt{N_D}$. In order to fabricate devices with a well-defined tunnel resistance, the doping levels in the semiconductor should be very precisely controlled. MBE-growth of GaAs allows precise control of the doping levels up to 10^{18}cm^{-3} . Above this number, Si (the n-type dopant) starts to behave amphotERICALLY, i.e. it can also occupy an As-position in the GaAs lattice and strong self-compensation can occur.[15] As a result, a precise control of the doping level is more difficult and the nominal doping levels will have to be adapted based on the experimentally obtained resistance of the tunnel contact.

Two-dimensional spin-injection-detection device. The targeted all-electrical spin injection-detection device has a typical lateral structure with two narrow ferromagnetic electrodes separated by a semiconductor channel, as shown in Fig. 9.3. We have used Medici 2D-simulations to optimize the doping level in these structures. The doping of the tunnel contacts is adopted from the 1D-simulations discussed above.

In the considered structure, the substrate is semi-insulating GaAs. The spin-transport region is a 150 nm wide n-type GaAs layer ($N_D = 5 \times$

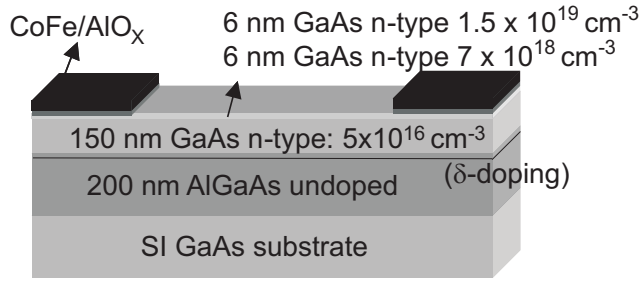


Figure 9.3: The device structure, in cross section.

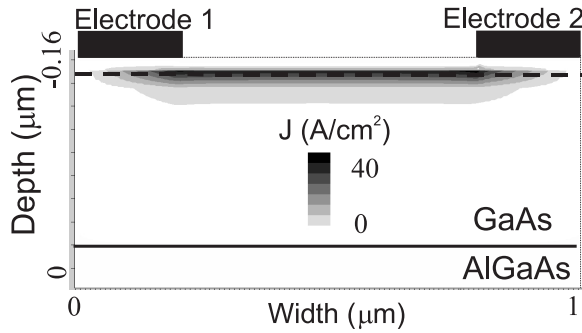


Figure 9.4: The 2D current characteristics of the spin-injection-detection device without recess etch. The region above the dashed line corresponds to the highly doped GaAs. In the semiconductor spin transport region the current is confined to the highly doped region near the surface. Note that the scale for the lateral and the vertical dimensions is different for clarity.

10^{16}cm^{-3}), with a 200 nm undoped AlGaAs cladding layer just below to confine the electrons in the GaAs layer. The Schottky tunnel barrier is generated by a thin, heavily doped n-type GaAs layer, with the layer structure adopted from the previous paragraph. On top of the structures, two electrodes are placed on an oxide tunnel barrier.

The doping level in the tunnel contacts is matched to a channel doping of $5 \times 10^{16}\text{cm}^{-3}$, which should result in long diffusion lengths. Hence, the device should be designed such that the current flows through a region with

the correct doping level. A simulation of the current distribution in this structure (Fig.9.4) shows that the major part of the current flows through the highly doped region at the surface. This results in both a reduced spin lifetime in the channel, and a mismatch between the channel and the contact resistances. A small recess etch can be used to remove the surface channel, however, our simulations show that when the highly doped GaAs is completely removed, the transport channel becomes fully depleted. An optimal recess etch would require nanometer precision in the etching process, which makes this structure too critical for practical applications.

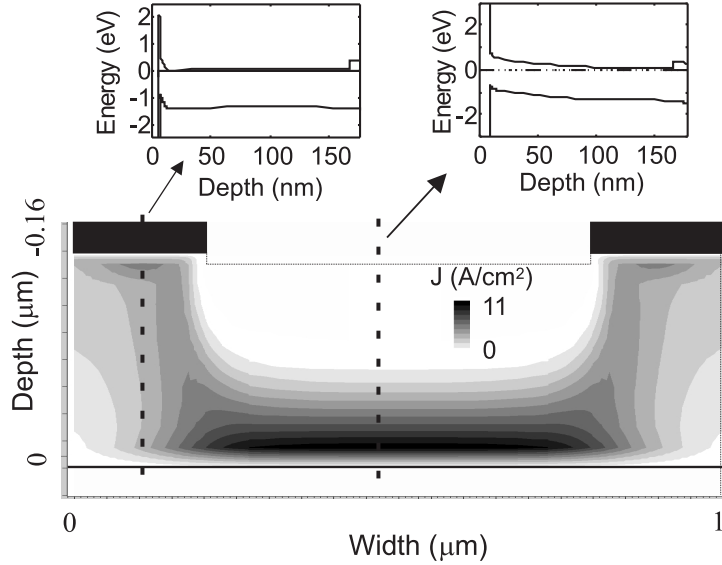


Figure 9.5: The 2D current characteristics of the spin-injection-detection device with a small recess etch (6 nm) and a δ -doped layer 10 nm below the AlGaAs/GaAs-interface. In the semiconductor spin transport region the current is confined in a region of about 40 nm near the AlGaAs/GaAs interface. The insets show the band profiles of the structure cross-sections at the metal contact and at the spin-transport region.

A more robust design, insensitive to etching variability, is obtained by inserting a delta-doping layer of $2 \times 10^{11} \text{cm}^{-2}$ close to the top of the AlGaAs cladding layer. This results in an inverse HEMT (High electron mobility transistor) structure that is immune to surface depletion effects. In Fig. 9.5, the current distribution is shown in this structure, with a 6 nm recess etch to remove the surface channel. The current in the spin-transport region

clearly flows close to the AlGaAs/GaAs-interface, where the carrier density is about $5 \times 10^{16} \text{ cm}^{-3}$. The insets show the band profiles of the structure cross-section at the position of the metal electrode and in the spin-transport region. From the band profiles it follows that underneath the metal contacts there is a more or less flat-band situation, which allows the injected electrons to be transported vertically to the channel near the AlGaAs/GaAs interface, where they are transported to the other electrode.

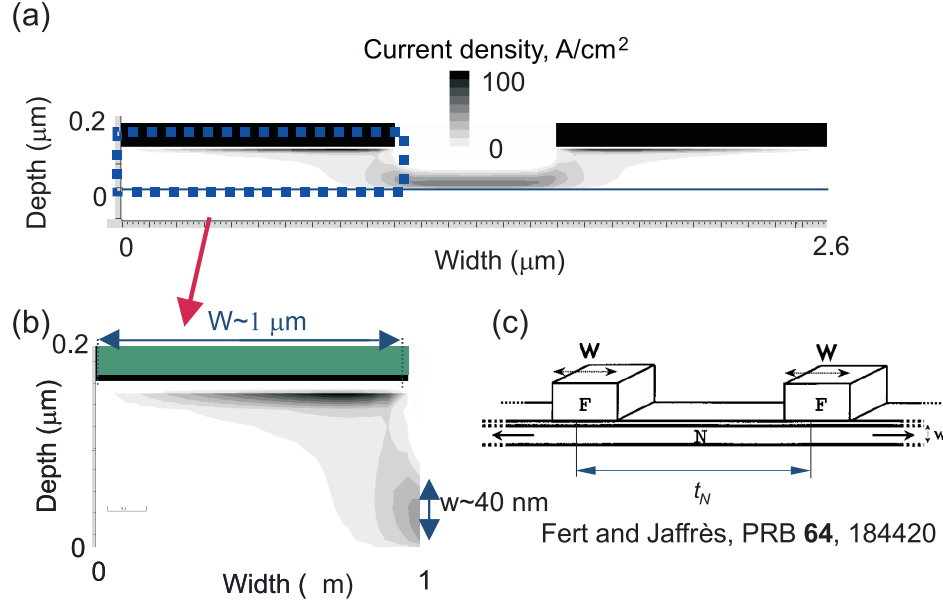


Figure 9.6: (a) The lateral current flow through the device using ferromagnetic electrodes with a width of $1 \mu\text{m}$. (b) A magnification of the current flow near the left metal contact. It is clear that the channel width, i.e. the effective width of the semiconductor channel is only 40 nm wide. (c) The diagram used by Fert and Jaffrès to calculate the magnetoresistance in a 2D-device.

The two-dimensional nature of the current path may change the resistance matching between the ferromagnetic metal and the semiconductor. Fert and Jaffrès[9] calculated the magnitude of the interface resistance in the case of a two-dimensional system, as depicted in Fig. 9.6(c). The optimal barrier resistance is given by the following equation:

$$r_N \left(t_N / l_{SF}^N \right)^2 (W/w) \ll r_b \ll r_N (W/w), \quad (9.1)$$

with r_N , the product of the resistivity with l_{SF}^N , the spin-diffusion length of the semiconductor, r_b the barrier resistance, W the width of the electrode, w the width of the channel and t_N the channel length. In order to be able to use their calculations, we need to extract the relevant parameters, as the effective width of the injector electrode and the semiconductor channel from the simulations. In the experimental situation, which will be described in the next paragraph, the ferromagnetic electrodes will have a width of $1\text{ }\mu\text{m}$. From Fig. 9.6(a), where the lateral current flow through the device is shown, it is clear that the channel width can be determined to be 40 nm . Using the equations of Fert and Jaffrès (eq. 9.1) we can estimate a number for the interface resistance: $W/w = 25$, $t_N = 1\text{ }\mu\text{m}$, $l_{SF}^N = 2\text{ }\mu\text{m}$, hence: $2.5 \times 10^{-8}\Omega\text{m}^2 \ll r_b \ll 1 \times 10^{-7}\Omega\text{m}^2$.

The simulations on Fig. 9.6(b) show that beneath the contacts the current is confined close to the surface, in the highly doped region. This can cause an enhanced spin relaxation of the injected carriers and a worse matching of the channel and the contact resistance. Reducing the contact width (as in Fig. 9.5), reduces the time that the injected electrons spend in the highly doped region and should hence increase the expected magnetoresistance effect.

9.5 Process flow

In the previous section we have designed the semiconductor structure to ensure an optimal current flow with respect to the carrier concentration in the different regions of the device. In order to practically realize all electrical spin injection-detection devices, there are several requirements concerning the actual device processing that have to be fulfilled.

The first requirement is set by the measurement geometry. As indicated in Chapter 2, the non-local geometry is virtually free of artifacts and allows an unambiguous proof of electrical spin injection and detection. For clarity, this geometry is schematically shown in Fig. 9.7. As a result, for the demonstration of electrical detection of electrically injected electron spins, this geometry is preferable from a scientific point of view. From an application point of view, a magneto-resistance-device in a two-terminal configuration is the first choice. In the non-local geometry, spins are injected in a channel and transported to a non-magnetic contact, while the potential difference between a ferromagnetic electrode that is within the spin diffusion length of the injector electrode, but outside of the current path and another non-magnetic contact to the channel is monitored as function of the applied magnetic field. Hence, we need two ferromagnetic contacts in close proximity of each other and two non-magnetic (ohmic) contacts to the channel.

In order to be able to measure the device resistance or the non-local signal of the device in a parallel and an anti-parallel alignment of the magnetization of the different electrodes, the two electrodes should exhibit different switching fields. The use of different materials or electrodes with a different geometry allows this. At the edges of long, thin ferromagnetic electrodes, typically flux closure domains occur. One should avoid that current is injected in the semiconductor from these edge domains, since the injected spin polarization will decrease. Furthermore, planarity is an important issue, since vertical steps in the electrode can generate high fringing fields, generating background signals such as the local Hall effect.

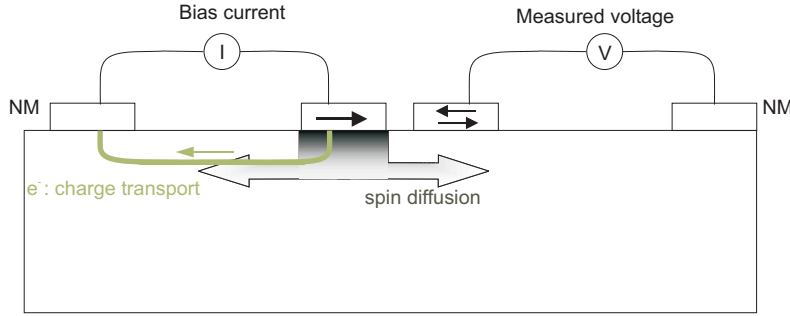


Figure 9.7: The non-local measurement geometry schematically shown. The charge current is driven from the injector electrode to a non-magnetic electrode while the induced spin accumulation diffuses also to the other ferromagnetic electrode that probes the splitting of the spin-dependent electrochemical potential near the interface with the semiconductor.

Finally, it is important that the processing leaves the tunnel barrier intact. AlO_X is known to be amphoteric, and can hence be removed by both acids and bases. Unfortunately, the solution to develop the optical resist contains KOH, a base, which can attack AlO_X . Hence, a protective layer should be added on top of AlO_X during processing. We have chosen for Ge as a protective layer since it protects the oxide from removal and can easily be removed prior to the metal deposition by a dip in H_2O_2 .

These arguments led to the following processing scheme, which is also schematically shown in figure 9.8:

In Fig. 9.8(top) the top view of the eventual structure is shown. There are four electrodes, the lateral ones connecting the semiconductor channel, the vertical ones connecting the ferromagnetic electrodes. The ferromagnetic electrodes have different shapes, allowing a different switching behaviour. Both electrodes are designed as a narrow stripe, 1 μm wide and

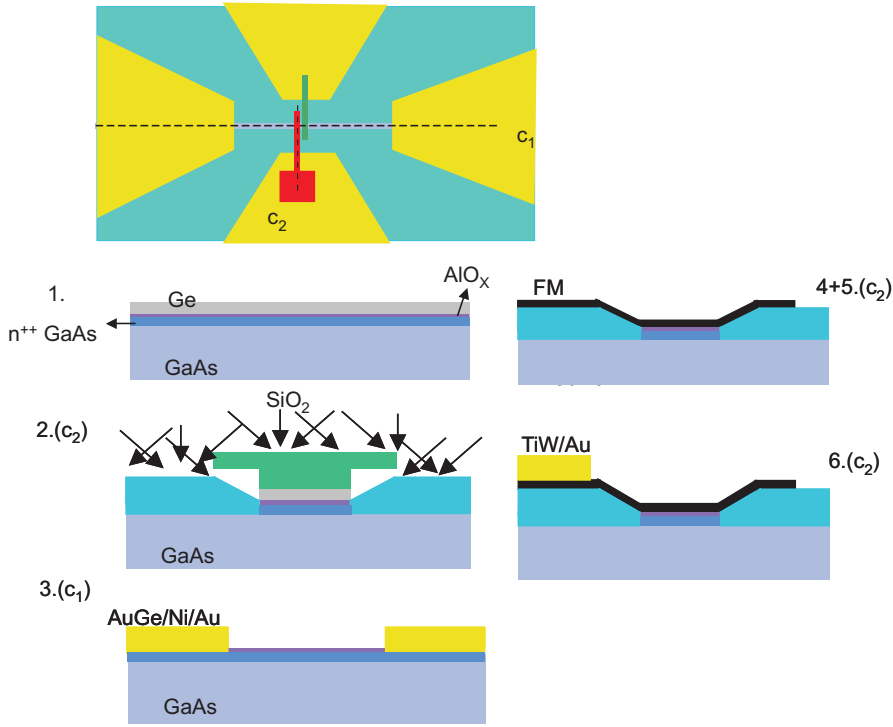


Figure 9.8: The processing scheme for the fabrication of the all-electrical spin-injection-detection devices. On top the final top view of the device is shown, with dashed lines, indicating two cross-sections. The different processing steps are shown below for the two cross-sections as indicated in the top figure: (c1): a cross-section parallel to the channel and (c2): a cross-section parallel to the ferromagnetic electrodes.

20 μm long. One of the two, however, has a larger square connected to it. The magnetization of this larger square reverses at lower values of the external magnetic field and pushes a domain wall into the narrow stripe.[16] This results in a smaller coercivity for this electrode. Furthermore, different materials are chosen for the two electrodes. The electrode with the square consists mainly of NiFe, while the other one consists of CoFe. NiFe is known to be a softer switching material. The ferromagnetic electrodes are deposited on top of insulating SiO_2 , and only touch the semiconductor in the channel, such that electrons are only injected from a selected part of the ferromagnetic electrodes, with a well-defined magnetization.

After the growth and the deposition of the AlO_x in the sputter system

using the same recipe as the one used to fabricate the AlO_x -based spin-LEDs, the sample is taken out of the sputter system and covered by a thin layer of Ge (20 nm), as indicated in Fig. 9.8(1).

Following the Ge deposition, a mesa-etch is performed by ion-milling and SiO_2 is sputtered in order to isolate the different mesas from each other. A double layer resist is spun onto the sample and developed. As depicted in Fig. 9.8(2), the developed double layer resist profile has a mushroom-like shape. Since sputtering is a deposition technique that is rather isotropic with respect to the angles of the incident atoms, this profile creates a smooth transition of the SiO_2 towards the semiconductor channel, avoiding large vertical steps for the ferromagnetic electrodes. After the SiO_2 -deposition, the structures are defined by lift-off. Prior to the SiO_2 deposition a short wet etch is performed to remove the highly doped region outside of the mesa.

In a third step, the metal connecting the semiconductor channel is deposited (Fig. 9.8(3)). Since we need an ohmic contact, we have chosen for a AuGe/Ni/Au-stack, which after annealing is known to provide a low contact resistance to n-type GaAs. Prior to the deposition, the Ge-layer is removed and a short HCl-dip is performed to remove the oxide on the GaAs-wafer. After the deposition and the lift-off, the structure is annealed in a N_2 -environment for 3 minutes at 370°C and 1 minute at 450°C .

In a fourth and fifth step, the ferromagnetic electrodes are deposited by means of lift-off (Fig. 9.8(4+5)). For the electrode with the square, 3 nm CoFe/42 nm NiFe/ 5 nm Ta is sputtered, while for the other one 45 nm CoFe/ 5 nm Ta is sputtered. The Ta-layer protects the underlaying metal from oxidation and the thin CoFe layer in the first electrode results in a high spin-polarization at the oxide/metal interface, while the magnetic switching behaviour is dominated by the thicker NiFe film on top. Prior to the deposition, the Ge-protecting layer is removed. The ferromagnetic electrodes are defined in different lithography steps. On the mask, the distance between the electrodes is varied for different mesas. Using this technique, we can fabricate devices with sub-micron spacing between the two electrodes using optical lithography. Furthermore, this step can easily be transferred to e-beam-lithography, allowing much smaller dimensions.

Finally the metal stack (TiW/Au) connecting the ferromagnetic electrodes is defined (Fig. 9.8(6)) by means of lift-off.

9.6 Initial experimental results.

9.6.1 Contact characterization

At this stage, no device has been finished using the complete desired processing flow. Hence, no magnetoresistance effects can be reported yet. However, some control experiments have been performed. In Fig. 9.9 the current-voltage and the conductance curves are shown for the device between one ferromagnetic electrode and an ohmic contact to the GaAs channel. The parabolic shape of the conductance vs voltage curve and the non-zero conductance at zero voltage indicates a tunneling process. Furthermore, the resistance at $V=0$ V equals $R_0 = 1.6 \times 10^{-8} \Omega$, which is just below the window of the desired barrier resistance as calculated using the equation from Fert and Jaffrès. The resistance can be tuned to fit the optimal resistance by changing the oxide thickness or the doping level in the semiconductor. Another possibility is reducing the width of the contacts. Indeed, as can be derived from eq. 9.1, reducing the contact width shifts the resistance window down.

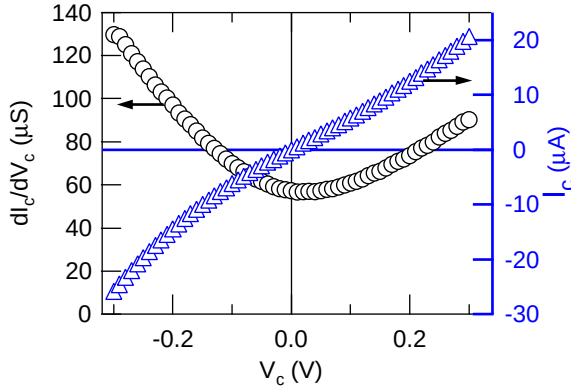


Figure 9.9: The current-voltage characteristics for one electrode and the associated dI/dV -curve at room temperature. The parabolic nature of the dI/dV and the non-zero zero-voltage conductance strongly indicates a tunneling process.

The temperature dependence of the resistance is an additional proof for tunneling as the dominant transport process. The tunnel process - as opposed to thermally assisted transport processes, is only weakly dependent on temperature. In Fig. 9.10 the resistance of the device is shown as function of the temperature. The resistance decreases slightly with increasing

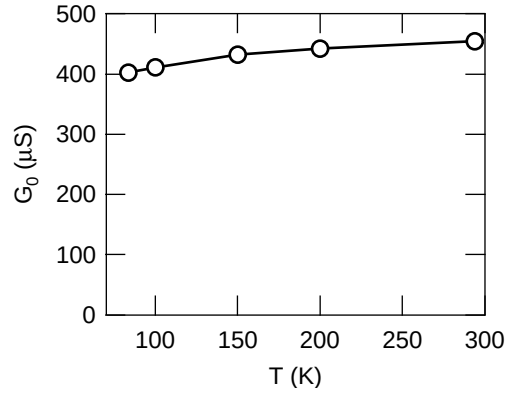


Figure 9.10: The resistance for one electrode-GaAs contact at zero volt as function of temperature.

temperature, consistent with tunneling behaviour.

9.6.2 Magnetic characterization

It is a necessity for operation of an all-electrical spin-injection-detection device that there is a magnetic field window where an anti-parallel and a parallel alignment of the magnetizations of the different ferromagnetic electrodes occurs. One can examine the magnetic behaviour of ferromagnetic electrodes by measuring their electrical resistance as function of the external magnetic field.[17] The resistance of a ferromagnetic metal depends on the angle between the current and the magnetization $\mathbf{M}$. This effect is called anisotropic magnetoresistance (AMR). The resistance is typically larger in the situation where $\mathbf{M}$ is parallel to the direction of the current than in the situation where it is perpendicular to the direction of the current. This effect can hence be employed as a sensitive tool for measuring the reversal of ferromagnetic electrodes. In order to examine the magnetic behaviour of the ferromagnetic electrodes, some have been contacted at both sides and the anisotropic magnetoresistance behaviour has been monitored. In Fig. 9.11 measurements are shown for both electrodes and it is clear that they exhibit a different switching field. Hence, both parallel and anti-parallel alignment of the ferromagnetic electrodes can be achieved.

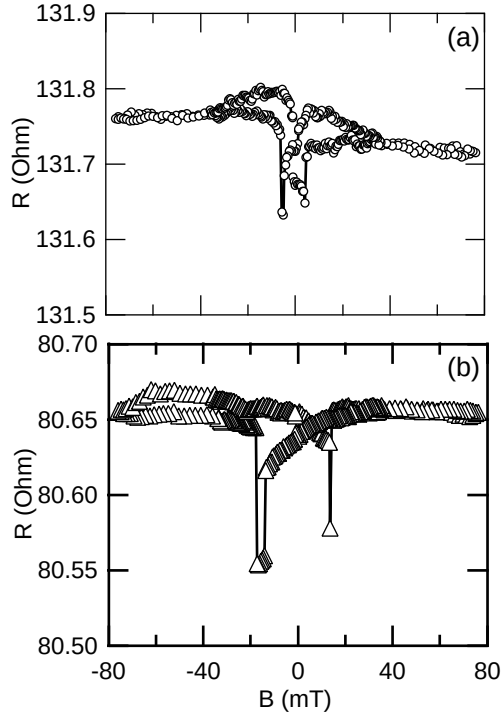


Figure 9.11: The AMR-curves for the electrode with (a) and without (b) square at room temperature. The difference in the switching fields is clear.

9.7 Conclusions

In summary, we have shown, using self-consistent simulations, that all-electrical spin-injection-detection devices require a specific design of the doping levels, both in the tunnel contacts and in the spin-transport region. The tunnel resistance is tuned by means of tailoring the doping levels in the tunnel contacts in order to maximize the achievable magneto-resistance, based on theoretical predictions. The doping level in the spin-transport region is optimized in order to have an optimal current distribution through the device. Furthermore, we have developed a processing scheme to fabricate these devices and initial experiments regarding contact characterization and magnetic characterization of the contacts have been performed.

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Chapter 10

Conclusions and further outlook

10.1 Conclusions

In the field of spintronics the spin is used as information carrier in addition to the charge. A lot of progress has been made already in metal-based spin-devices, with applications in hard-disk read-heads and Magnetic RAM. A further integration of the spin in conventional electronics would implicate the use of spin in semiconductor devices. The first prerequisite for such devices is the successful creation of a non-equilibrium spin polarization in semiconductors. The most straightforward approach is direct injection of spins from ferromagnetic materials. This has proven to be non-trivial due to the large difference in the conductance between metals and semiconductors. However, introducing a tunnel barrier between the ferromagnetic metal and the semiconductor bypasses this issue.

In this work we have explored a variety of tunneling contacts to demonstrate electrical spin injection in GaAs. A measurement of the circular polarization of the light emitted in a light-emitting diode has been employed as a reliable tool to measure the injected spin polarization. Using the oblique Hanle effect, a technique which relies on the precession of the injected spins in an oblique magnetic field, both the magnitude of the injected spin polarization and information about the spin dynamics can be extracted from the field-dependence of the circular polarization of the light. In order to demonstrate and study spin injection in semiconductor heterostructures, spin-light-emitting diodes have been designed and fabricated using different injector contacts.

Spin-LEDs based on tunnel injection from a sputtered CoFe ferromag-

netic metal through an AlO_x -barrier have been shown to result in a high injected spin polarization that persisted up to room temperature. Furthermore we have examined and explained the bias dependence of the injected spin polarization. We have supported our interpretation using self-consistent simulations.

Spin-LEDs based on tunnel injection from an epitaxially grown MnSb ferromagnetic metal through a tailored, narrow Schottky-barrier have been fabricated and shown to result in spin injection at low temperatures. Using self-consistent simulations we explained the strong bias dependence in this type of spin-LED.

Another route that we explored was spin injection from the hole-based ferromagnetic semiconductor (Ga,Mn)As, which has been predicted to exhibit a half-metallic nature (i.e. 100% spin-polarized). Indeed, we have shown a very high injected spin polarization of 80% at liquid Helium temperature. This effect depends furthermore strongly on the applied bias, which allows voltage controlled spin-injection. We have explained the strong bias dependence of the injected spin polarization using a combination of self-consistent simulations of the charge distribution and the band bending and a model of ballistic tunneling using the Landauer-Büttiker formalism based on a band-structure calculated by a tight-binding model. The latter calculations were performed in a collaboration with a theoretical physics group in Warsaw.

Apart from a characterization of the different injector structures based on their performance as a spin-injector, we looked into the electrical characteristics of the injectors. We were able to show that the oxide injectors behave different from the Schottky-barrier and the interband-tunneling type of injectors in terms of band-alignment and parasitic hole currents. Furthermore we were able to distinguish a mechanism that can create additional unpolarized electrons at sufficient bias, impact ionization.

Our measurement geometry, which relies on the precession of the injected spins made it possible to examine the electronic-nuclear interactions in the GaAs-layer. The hyperfine induced nuclear spin polarization acts on the electrons as an effective field, accelerating the spin precession, which is directly probed by the oblique Hanle effect. We have shown intriguing transient effects, corresponding to the coupling between the electrons and the nuclei. Using alternating magnetic fields we were also able to resonantly depolarize the nuclear spins. Furthermore, we examined and explained the transient effects, the zero-field depolarization of the nuclear spins and the resonant depolarization of the different isotopes.

Finally, we have looked into the design of all-electrical spin injection-detection devices. Using self-consistent simulations, we have designed the doping profile for both the injection contacts and for the spin-transport region. The tunnel resistance is tuned by means of tailoring the doping

levels in the tunnel contacts in order to maximize the achievable magnetoresistance, based on theoretical predictions. The doping level in the spin-transport region is optimized in order to have an optimal current distribution through the device. Furthermore, we have developed a processing scheme to fabricate these devices and initial experiments regarding contact characterization and magnetic characterization of the contacts have been performed.

10.2 Outlook

In this thesis it has been demonstrated that different types of injectors can result in efficient electron spin injection. Meanwhile different groups throughout the world have confirmed and partially extended these results, indicating that electrical spin injection in semiconductors can be achieved by an appropriate design and fabrication of the injector structure, taking into account both the ferromagnetic injector and the semiconductor below.

Spin-LEDs are interesting proof-of-principle tools to show electrical spin injection and the spin dynamics in direct-band semiconductors. It is obvious that optical detection in a spin-LED of spin injection in indirect-band semiconductor such as Si and Ge is merely impossible. Hence, for these materials an electrical detection of a spin ensemble in semiconductors is desirable. Furthermore, also from an application point of view, all-electrical spin-injection and detection devices are better candidates for applications. At this point, no reports have been published that unambiguously show such an accomplishment. As to realize all-electrical spin-injection and detection, a careful design and fabrication of both the semiconductor part and the ferromagnetic injector part, as discussed in Chapter 9, is necessary.

Electrical spin injection in Si seems the next big hurdle to be taken. The success of its interstates-free native oxide, SiO_2 , seems to indicate that SiO_2 is the logical choice for the tunnel junction. However, as indicated in Chapter 9, the band-alignment of the ferromagnetic metal and the semiconductor is of great importance to realize tunnel injection. Since Fermi level pinning does not occur at the SiO_2 -Si interface, the metal work function determines the band-alignment. With the lack of low-work function metals that are ferromagnetic at room temperature, one needs to realize narrow heavily doped layers near the oxide-semiconductor interface. Although Si processing has a rich history, this still is a challenging task. Transferring technology developed on III-V-semiconductors to Si, brings of course new problems, new challenges and new possibilities.

A full integration of magnetism and charge-based electronics is realized in ferromagnetic semiconductors. In this work, we were able to show that highly efficient spin injection can be realized in fully semiconductor struc-

tures, using a p-type ferromagnetic semiconductor as spin source. Since electron injection is preferred over hole injection, interband tunneling can lead to electron spin injection in non-magnetic semiconductors. However, the relatively low Curie temperature limits still the applicability of this material system. Different groups are investigating ferromagnetic semiconductors and try to reach stable room temperature operation. If this can be realized, there is a bright future for ferromagnetic semiconductors.

We were able to show that electrical spin injection can lead to fascinating interactions with the host nuclear spins of the semiconductor lattice. The long lifetime of the nuclear spins makes it a quantum system suitable to function as a quantum bit in quantum computers. Electrical spin injection may be used in this case to initialize and/or to read the quantum state prior to and after the calculation.

Concluding, the quest for complete spintronic semiconductor devices, relying on the spin rather than on the charge, is not completed yet, but one hurdle definitely has been taken. We hope that this study could contribute to the understanding of the injection process and the transport properties for different types of injectors.