that the base can contain fewer layers (no seed layers, no bonding layer). Disadvantages are the smaller emitter current compared to a Schottky launcher, and the unknown spin-dependent transmission characteristics of our ferromagnetic films. (Most of them are Mn-based compound ferromagnets such as MnSb, MnAs, MnAl, MnGa, and NiMnSb. However, in December 2003 an Fe source has been installed on the system).

Initial growth and processing tests have been performed for MnSb and NiMnSb based devices, but finished devices have not yet been made. This work will be described in more detail in a later report.

**Introduction and issues:**

The main task of UT in WP1 is to use hot-electron spin filtering in standard ferromagnetic materials for spin-injection into semiconductors (see figg. b15). In this approach, we use a three-terminal transistor structure (spin-valve transistor, SVT) with a hot-electron emitter, a ferromagnetic metal base and a semiconductor collector. Such structures are routinely fabricated in our lab using vacuum metal bonding with Si as both emitter and collector. However, some important changes have to be made to directly measure the spin polarization in the collector.

In order to detect the polarization of the spins in the semiconductor, a Light Emitting Diode (LED) is used as the collector of the transistor. These structures are supplied by partner IMEC. The LED consists of a p-GaAs substrate, a p-type AlGaAs (30% Al) blocking layer, an active layer of 100 nm GaAs, and a surface layer of 40 nm AlGaAs (10% or 20% Al). The blocking layer is to confine the injected electrons in the active GaAs region where radiative recombination takes place, from which the spin-polarization can be determined using the Oblique Hanle effect (as is well established by the IMEC group). The incorporation of this LED collector structure raises the following issues:

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**Fig. b15:** (a) Detailed band diagram of the structure designed for hot electron spin-injection and detection using an AlGaAs LED. (b) Similar, but then under application of emitter bias, and a bias between base and collector.
1) **Hole current**

The LED collector is p-type to supply holes for recombination. Traditionally an n-type Si collector was used, providing the Schottky barrier for the necessary energy and momentum selection. While the LED structure also forms a barrier with the metal base, small forward bias is needed to lower the energy band bending. The bias creates a flow of holes (the majority carriers) towards the metal base, and thus a current in the same direction as the current of spin-polarized electrons injected into the collector from the base. This hole current therefore complicates unambiguous electrical measurement of the injected hot-electron current, although it does not necessarily affect the luminescence process. This issue is particularly critical for achieving RT operation, where hole currents are larger, as shown below.

2) **Emitter structure**

The LED collector is expected to create a collector barrier of approximately 1 eV, independent of base metal, due to Fermi level pinning. Such barrier height is larger than any emitter Schottky barrier that can be created on Si (highest is Si/Pt barrier contact of 0.9eV). Thus, the traditional Si Schottky emitter cannot be used, as the emitted hot electrons do not have sufficient energy to enter the LED collector.

3) **Bonding**

The surface roughness of the LED as well as the emitter structure needs to be sufficiently low for the vacuum metal bonding to work.

**Results:**

**Results 1) Hole current**

To address this issue, LED structures having an AlGaAs surface layer were fabricated. At 10% Al content, the bandgap of AlGaAs is 1.55 eV, thus 0.13 eV higher than that of the active GaAs layer. This creates a step in the valence band of about 0.08 eV to reduce the hole flow into the metal base. To test this, Au-LED diodes were fabricated by evaporation of Au onto pre-cleaned pieces of LED wafer. These were patterned into 350x750 micron$^2$ areas using standard photolithography and ion beam etching (IBE). I-V-measurements (fig b16) show clear diode characteristics in the studied temperature range (100 K - 300 K).

In the full structure shown in fig b15, a forward bias voltage of about 0.4 V across the base-LED diode is required to lower the band bending in the LED. At room temperature we found that the resulting hole current into the Au metal is too large, typically in the mA range. This is much larger than the expected current of injected hot electrons (typically microamperes (mA)). However, as shown in fig b16, the hole current at 100 K is reduced to an acceptable level, on the order of 10-12 mA at a forward bias of 0.4 V. To further improve this, we fabricated a second set of LED diodes with larger Al concentration (20%) in the surface layer. At higher Al content, the bandgap of the AlGaAs layer is larger, and thus the step in the valence band is increased. For the diodes on LED’s with 20% Al content, the hole current at 0.4 V was about 4 mA (see fig.), which is an improvement by a factor of 3 as compared to the LEDs with 10%
Al surface layer. For the low temperature demonstration (Deliverable D7, WP1 (month 24)) this was sufficient, noting that we have recently demonstrated hot electron collector currents above 40 mA in Si based devices. To achieve room temperature operation it is desirable to further reduce the hole current, or use a lock-in detection technique were the injected hot-electron current is modulated using the emitter current, while the base-collector LED bias that produces the hole current is kept constant.

**Results 2) Emitter structure**

To address this issue, the option of using alternative semiconductor materials (such as SiC or GaN) that allow higher emitter Schottky barriers above 1 eV was considered. This option was dismissed, as it would involve too many modifications in the fabrication technologies. The second option that was considered was the use of a Metal-Insulator-Metal (MIM) structure as the emitter, as is commonly used in the magnetic tunnel transistor. Such structure raised a concern, at least at this moment, on the stability of the thin tunnel barrier under the required high bias and high emitter current. We therefore decided to use a novel type of emitter structure, the MIS diode (Metal-Insulator-Semiconductor). This allows one to obtain high injection energy by applying a high bias voltage that drops partly across the insulator and partly on the band bending region of the semiconductor. Also, we anticipated that the use of MIS diodes with Si would allow us to use most of the existing device process scheme.

**Fabrication and characterization of MIS diodes:**

Several MIS diodes with sizes between 10 and 180 micron have been fabricated and characterized electrically, using different metals. As insulator Al2O3 as well as SiO2 was investigated. The Al2O3 was prepared by evaporation of 0.7 nm of Al onto HF treated Si.

![Fig. b16: Current voltage characteristics of Au/LED structures with 10% and 20% Al content in the surface layer.](image-url)
substrates, and subsequent in-situ natural oxidation in pure oxygen gas. This results in a Al2O3 tunnel barrier of about 1 nm. To obtain thicker Al2O3 barriers, this procedure was repeated once more. Diodes with SiOx insulator were prepared by in-situ natural oxidation of HF treated Si. All oxidations were done at room temperature. The diode characteristics were compared to diodes without the insulator.

High quality diodes were successfully created. The I-V measurements (fig b17) on diodes with about 1 nm of Al2O3 show that the barrier height depends on the type of metal electrode, as expected for diodes with a low density of interface states at the Insulator-Semiconductor boundary. Values for the barrier height (at zero bias) are 0.8 eV for Au, about 0.72 eV for Co and 0.64 eV for NiFe electrodes. Characteristics of diodes with different insulators are shown in fig. b18. When compared to diodes without insulator, the MIS diodes with 1 nm of Al2O3 or with SiOx show similar I-V-characteristics, indicating that the tunnel barrier is electrically transparent. However, for diodes with 2 nm Al2O3 barrier a clear reduction of forward and reverse bias current was observed, corresponding to an effective barrier height of 0.9 eV at zero bias for Au/Al2O3/Si diodes. Moreover, the ideality factor increased as compared to Au/ Si Schottky diodes. These results indicate that the insulator limits the current and part of the bias voltage drops across the insulator, as desired. The results support the use of such MIS diodes as emitters of hot electrons with energies of 1 eV and beyond.

Integration of the MIS diode in the spin-valve transistor:

Next, we decided first to test whether these MIS diodes are suitable to act as hot-electron emitter in the spin-valve transistor, and specifically, whether the expected increase of hot-
electron energy can be achieved. Thus we have successfully fabricated a novel spin-valve transistor with MIS injector. The typical device has the following structure from emitter to collector: Si / Al2O3 / Co / Au / NiFe / Au / Si. The vacuum metal bonding is done using the last Au layer. Compared to the conventional spin-valve transistor, the introduction of the MIS on the emitter side allows tuning of the energy of the hot-electrons injected into the base. This configuration allows higher emitter current $I_e$ and hot-electron energy, which is beneficial for the output current. The characteristics of this device are presented in the following to demonstrate the capability of MIS hot-electron injector and its advantage over conventional Schottky diode and MIM injectors.

The principal parameters of an SVT with MIS injector are shown in Fig. b19 and b20. The first figure b19 shows the dependence of collector current, magnetocurrent (MC) and transfer ratio $I_C/I_E$ on emitter current. Fig. b20 displays the magnetic response of the device i.e. the dependence of collector current on the applied magnetic field. We observe a steeply enhanced collector current when $I_e$ is increased (so far up to 200 mA) as it was expected for a SVT device with MIS injector. For comparison, we note that for SVT devices without the MIS injector, the collector current is simply linearly proportional to the emitter current, such that the transfer ratio $I_C/I_E$ is independent of emitter current. This is due to the fact that the energy of hot electrons injected across a Schottky diode is determined by the Schottky barrier height, which remains constant. For the device with MIS injector, we see a marked increase of the transfer ratio, proving that the hot electron energy increases with increasing emitter current. Also note that at low emitter bias, the barrier height for the Si/Al2O3/Co MIS emitter (0.72 eV) is lower than that of the Au/Si collector Schottky barrier (0.8 eV). This explains why
hardly any collector current is obtained at small emitter current. Altogether, the electrical behavior establishes the principle of operation of the MIS injector i.e. part of the emitter bias drops across the insulator such that the energy of the hot electrons increases with emitter bias. Interestingly, with increasing $I_e$, the magnetocurrent of the SVT with MIS injector also grows to values of 160%, in contrast to the behavior that is generally observed for the SVT or magnetic tunnel transistor. The origin of this effect is under further investigation.

**Fabrication issues for transistor with MIS diode:**

The fabrication of the MIS-SVT encountered a few unexpected difficulties, which resulted in some delay. The introduction of the aluminum oxide in the structure brought on two

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Fig. b19: Collector current $I_C$ for parallel and antiparallel state of the spin-valve base (top panel), relative magnetocurrent $MC$ (middle) and transfer ratio $I_C/I_E$ (bottom) as a function of emitter current at 90K, for a MIS-SVT with structure Si / Al2O3 / Co / Au / NiFe / Au / Si.
technological issues, which were (i) the survival of AlOx in the TMAH (Tetra-Methyl-Ammonium Hydroxide) etching solution, and (ii) the adhesion between AlOx and Au. These are discussed in more detail next.

In the standard fabrication process for the SVT, we use TMAH as the etching solution for the removal of the silicon handle wafer part of the SOI wafer, which acts as the silicon emitter. This etching has always been carried out with 10% wt TMAH solution at 85°C resulting in an etch rate of 0.6 to 1.0 um/min. The etching time for complete removal of the silicon therefore requires submergence of the full bonded structure in TMAH for more than 6 hours. Repeated experiments showed that the stacks containing the MIS emitter failed to survive more than 3 hours. The AlOx turned out to be the cause of trouble. We thus had to look for an alternative method for thinning the emitter wafer. Cryogenic plasma etching was then proposed and employed for silicon etching with prominent advantages in time, controllability, reproducibility.

The interface between the AlOx and the first metal (Au) in the base turned out to be another problem, causing bonded structures to detach at the AlOx/Au interface during processing of the devices. The energy of adhesion of these two materials is relatively low (265 mJ/m2) compared to 1140 mJ/m2 of Co/AlOx. Thus, it was decided to temporarily omit the Au layer and place Co in direct contact with AlOx. This means the effective emitter barrier height would be reduced. Nevertheless, these modifications proved to work as the first MIS-SVT was realized and presented above.

**Results 3) Bonding.**

To address issue 3, Transmission electron microscopy (TEM) on the surface of the MIS injector was performed. The cross-sectional images show that the metal grown on top of the
MIS diodes is smooth, and that the roughness complies well with the roughness requirement for successful vacuum metal bonding. Then, also the surfaces of the LED wafers supplied by IMEC were investigated by Atomic Force Microscopy (AFM). We found that the LED surface is rougher at higher content of aluminum in GaAs, although AFM investigation of 10% and 20% Al content surface reveals only minor differences in the center of the wafer. Larger roughness was however observed in different locations on the same substrate, so the uniformity needs to be improved. This is done by partner IMEC.

The native oxide on the AlGaAs surface could potentially be a roughening factor and require a chemical removal prior to any device processing. A near isotropic chemical etching using NH4OH:H2O2:H2O for particles and native oxide removal of AlGaAs has been developed.

The first test on vacuum metal bonding between a MIS injector and LED collector was successful, indicating that the roughness levels are sufficiently low. Unfortunately, the structure did not survive the processing, because it contained an Au/AlOx interface with poor adhesion, a problem that was already discussed above.

**Discussion of the results and comparison with workplan:**

From the results presented above it is clear that the deliverable Deliverable D7, WP1 (month 24): "Demonstration of injection of highly spin polarized hot electrons into a semiconductor at low temperature" has not yet been achieved. However, it should be mentioned that all the issues concerning this goal have been addressed. The LED structure has been designed and metal-LED diodes that have sufficiently small hole current are obtained. Also, the problem of enhancing the hot-electron energy has been addressed by using a MIS injector, and it has been demonstrated that it works as expected (in passing, this has resulted in a new type of hot-electron transistor which may have advantages as compared to the existing SVT, while work on Si based MIS diodes may also prove useful for remaining parts of the project involving Si based spin injection devices). Finally, the roughness of the LED as well as of the MIS diodes was found to be sufficiently low, and first successful bonding of the complete structure was obtained.

As already mentioned above, there were two unexpected processing problems associated with the introduction of the Al2O3 insulator, it did not survive our standard etch in TMAH, and it has poor adhesion with Au. Of course, these problems with the Al2O3 caused delay because of the time involved in understanding what went wrong in the process, and in developing and testing the alternative process. However, at present these issues have been resolved and there is as of yet no fundamental reason why we should not be able to achieve the deliverable.

**Plan for the next year:**

Given the delay, our first priority for the coming year is to demonstrate the injection of highly spin polarized hot electrons into a semiconductor at low temperature (which is in fact Deliverable D7, WP1 (month 24).
The following activities are to be carried out:

- Fabrication and processing of a transistor device with MIS injector and LED collector, and a magnetic metal base.

- Electrical characterization of the device and determination of the magnitude of the hot-electron current arriving in the semiconductor LED.

- Measurement of the luminescence induced by the hot-electron current injected into the semiconductor LED, and determination of the spin polarization using the Oblique Hanle Effect at low temperature (in collaboration with IMEC).

We stress that for these activities, all the necessary ingredients have been developed, and there is no fundamental reason why this should not be achieved within the next few months. We should also mention that, based on the knowledge gained during the project so far, the step from low temperature operation to room temperature operation appears to be a relatively little one. Since we already know that hot-electron spin transport is quite insensitive to temperature in the traditional ferromagnetic materials (e.g. cobalt) we are using, the only issue seems to be the enhanced hole current in the LED collector. Although we expect that this is not a problem for the luminescence, a straightforward and simple solution was already identified (the use of a modulation technique). Therefore, we expect that we can still achieve the final Deliverable D13, WP1 (month 36) ”demonstrate the injection of highly spin polarized hot electrons into a semiconductor at room temperature“.

Nevertheless, an assessment of the progress will be made at the project meeting around month 30, and possible adjustments of the final milestone or refocusing of our efforts onto possibly more promising parts of the project will be discussed at that time.
**Part 2/2c: (WP1) Electrical measurements and spin injection into silicon**

**Introduction**

One of the main challenges of WP1 in year 2 of SPINOSA was the spin injection into silicon. One reason why the detection of spin injection into silicon is so difficult is that it is impossible to build (efficient) LEDs out of the indirect band gap material. So besides the approach based on Faraday rotation which was described in part a all detection experiments rely on purely electrical transport experiments. As will be described in the following a number of experiments have been carried out, none of them giving the indication of spin injection so far. However, a lot of optimization has been done and meanwhile high quality tunneling contacts on highly doped silicon can be fabricated and a lot more knowledge about side effects has been obtained. One experiment in which a tunnel contact between a magnetic semiconductor and a non-magnetic material exhibits a 10% spin valve signal (without any spin-valve) shows again, that extreme care has to be taken when the results of magnetotransport experiments are interpreted. However, based on the results obtained until the end of year 2 we stand a high chance to realize spin injection into silicon, unless major basic obstacles can be identified that prevent spin injection based on the existing technology.

**Tunnel barriers on (Ga,Mn)As**

As a first step for the experiments on tunnel barriers at UWUERZ the spin dependence of the tunneling process should be demonstrated. This can be done in a GMR like geometry where two ferromagnetic tunneling contacts are put on a semiconductor mesa. When spin polarized transport occurs between the contacts a difference in resistance should be detected when the magnets switch from parallel to antiparallel magnetization. However, if the electrodes

![Diagram of Co/AlOx/(Ga,Mn)As tunnel device]

*Fig. c1: Schematic diagram of Co/AlOx/(Ga,Mn)As tunnel device.*
are too far apart or other device parameters are unsuitable for the experiment it may occur that spin dependent tunneling occurs but no spin-valve signal can be detected. It seems thus suitable to demonstrate first the spin selectivity of the tunneling contact by preparing Co/AlOx contacts on a ferromagnetic semiconductor, resulting in a TMR geometry. In this device, the spin dependence can easily be detected because the contact resistance itself will change in the magnetic field and the tunnel barrier can be tuned to very high resistances, giving a large effect without the need of spin polarized transport through the semiconductor. We chose (Ga,Mn)As as a ferromagnetic semiconductor, because it is available in Würzburg and has been studied extensively over the last two years.

**Sample Fabrication:**

The samples were fabricated as follows.

a- A (Ga,Mn)As layer was grown on GaAs wafer with a thickness of 250nm in MBE chamber. The (Ga,Mn)As layer was highly doped (around $10^{19}$ atoms/cm$^3$).

b- The (Ga,Mn)As layer was moved to the sputtering chamber to Sputter a 1.4nm Al layer on top of (Ga,Mn)As layer.

c- The Al layer was oxidized in situ (inside the chamber) for 8 hours under pressure of 100 mbar to create an AlOx layer.

d- A 25nm cobalt layer was sputtered on the top of the AlOx.

e- At the end of the sputtering process the sample has the following vertical structure:

GaAs (substrate)/ 250nm (Ga,Mn)As/ 1.4nm AlOx/ 25nm Co.

The next step is to use optical lithography to define a tunneling device with dimension of 100nm x 100nm. After using optical lithography, the device was etched using CAIBE by Ar gas till reach the (Ga,Mn)As layer. The next lithography step is to make contact pads (300nm
Year 2
Part 2: Work progress overview

b. Electrical Measurements:

The current-voltage characteristic (I-V) of the device was measured using a probe station and a HP 4155 semiconductor parameter analyzer by applying voltage and measuring the current through the top layer (device) and the bottom layer (Ga,Mn)As. Figure 4.4 shows the I-V curve and the resistance of a Co/AlOx/GaMnAs device with dimensions of 100mm.

The I-V shows non-linear behavior which is similar to the theoretical I-V curves calculated by Simon (1963). This non-linearity is caused by a decrease in the effective barrier height with increased applied voltage. From this curve we conclude that a 1.4nm AlOx is sufficient for tunneling.

The difference between the two figures is the direction of the magnetization of the two layers with respect to the external magnetic field. In Figure 4.4, the easy axis of the magnetization of the two layers lies parallel to the external magnetic field, so the application of a small magnetic field is sufficient to switch the magnetization, while in Figure 4.5 the field is along the hard axis and must be larger before switching occurs.

Using SQUID, the magnetization of the three layers continuous film was measured as a function of external magnetic field. Measurements were performed with the field aligned either in hard or easy axis as shown in Figure 4.5a and 4.5b respectively.

Figure 4.3 shows the SQUID loops of a Co/AlOx/GaMnAs trilayer which shows two distinct switching fields. The first switching step in the magnetization curve corresponds to the Co layer, while the larger switching step corresponds to the Ga,MnAs layer. There are clearly identified by comparison to the switching of the single layer of Co and Ga,MnAs from previous measurements. The separate switching of the Co and the Ga,MnAs indicates that the AlOx barrier is pin hole free and thick enough to separate the switching of the two ferromagnetic materials.

The difference between the two figures is the direction of the magnetization of the two layers with respect to the external magnetic field. In Figure 4.5a, the easy axis of the magnetization of the two layers lies parallel to the external magnetic field, so the application of a small magnetic field is sufficient to switch the magnetization, while in Figure 4.5b the field is along the hard axis.

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The difference between the two figures is the direction of the magnetization of the two layers with respect to the external magnetic field. In Figure 4.5a, the easy axis of the magnetization of the two layers lies parallel to the external magnetic field, so a very small magnetic field is required to switch the magnetization, while in Figure 4.5b the field is along the hard axis and must be larger before switching occurs.

The I-V shows non-linear behavior which is similar to the theoretical I-V curves calculated by Simon (1963). This non-linearity is caused by a decrease in the effective barrier height with increased applied voltage. From this curve we conclude that a 1.4nm AlOx is sufficient for tunneling.

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thick to get tunnel barrier and is pin hole free. It is therefore appropriate for use for spin injection from a ferromagnetic into a semiconductor.

c. Magnetoresistance Measurements:

Figure c5 shows the resistance as a function of the external magnetic field at different applied voltages. At high positive magnetic field, the two magnetization are saturated and
with decreasing magnetic field the Co layer starts to rotate (switch) and the magnetization of the two layers become antiparallel, thus increasing the resistance until the magnetization of the (Ga,Mn)As rotates (switches) and the magnetization in the two layers become parallel again, leading to decrease the resistance. The measured resistance values for the parallel and antiparallel correspond to a magnetoresistance of 10% (at 600mV and at 4K). The magnetoresistance is higher than the resistance of a single (Ga,Mn)As/Co hybrid structure, so that any intrinsic magnetoresistance in the materials can be excluded. Moreover, the magnetoresistance scales with the resistance of the tunnel barrier. Although this seemed to be clear evidence for a TMR effect in a Co/AlOx/(Ga,Mn)As junction, more control experiments were performed that lead to a surprising result.

**d. A novel magnetoresistance effect**

For the control experiment a tunneling contact was fabricated in which the Co-layer was replaced by a gold layer. In this structure no TMR is to be expected, because only one side of the contact is ferromagnetic. However a field sweep revealed an effect which is similar in field dependence and magnitude as for the all magnetic sample. Fig. c6. Using a previously installed magnet cryostat which allows for continuous changing of the angle in which the magnetic field is applied with respect to the sample, several field sweeps under different angles were done. The results can be seen in Fig. c7. As can be seen, the features of the curve strongly depend on the orientation of the magnetic field and are similar to what has been observed in bulk (Ga,Mn)As by M. Roukes and coworkers (H.X. Tang et al. Phys. Rev. Lett. 90 (10), (2003) 107201) and which they mention as Giant Planar Hall effect. This effect is

![Graph](image-url)

*Fig. c6: Magnetoresistance curve of a Au/AlOx/(Ga,Mn)As sample. The field was applied in the plane of the GaMnAs layer. The giant planar Hall effect signal of the sample is amplified by the tunnel barrier and gives the exact signature of a spin valve.*
based on the anisotropic magnetoresistance in (Ga,Mn)As and appears when a magnetic field is applied in the plane of the sample. However, although in the tunneling structure the relative effect is similar to what has been observed in the bulk, the absolute effect is now approx. scaled by the factor $R_{TB}/R_{(Ga,Mn)As}$ where $R_{TB}$ is the resistance of the tunnel barrier and $R_{(Ga,Mn)As}$ is the resistance of the bulk (Ga,Mn)As sample. At the moment we are trying to explain the effect in collaboration with Thomas Jungwirth. One possible explanation is the change in the density of states in (Ga,Mn)As during remagnetization. Although the total density of states does not change, the k-dependence towards the tunnel barrier may vary and thus change the tunneling probability. In any case, it may be possible to fabricate devices which can be used as sensors for angular alignment and the possibility of taking a patent is considered.

**Tunnel barriers on silicon**

One of the main efforts of UWUERZ in the last year was dedicated to spin injection into silicon. The most promising approach in this material system is based on tunnel barriers between ferromagnetic metals and silicon. The most promising candidates for the tunnel barriers are silicon nitride, silicon oxide (due to their omnipresence in silicon technology and their high quality) but also AlOx due to its performance in TMR contacts and in spin injection experiments (IMEC) and the reliable deposition of thin high quality layers. The spin injection experiments in UWUERZ (that will be described later) have been using two different sets of samples, one fabricated in Wuerzburg and another set of reference samples that were fabricated at Twente. The reference samples were used because of several reasons. Firstly, a new sputtering system is setup at the monet in Wuerzburg which is not yet operational, so that all sputtering takes place in an old high vacuum machine. Secondly, UT has a long standing experience in the fabrication of tunnel barriers that can help in making high quality devices and allow for an
assessment of the quality of the tunnel barriers fabricated in Wuerzburg. All tunnel barriers investigated were fabricated on highly n-doped silicon wafers, in order to avoid carrier freeze-out at low temperatures and additional Schottky barriers that can form underneath tunnel barriers (see UT contribution on hot electron transport). In addition these wafers also allow for the formation of additional ohmic contacts without the need for alloying or ion implantation.

At UWUERZ the fabrication process for Co/AlOx/Si tunnel barriers was as follows:

a- A highly doped Si wafers (resistance varies between 0-1mW/cm) were cleaned with standard cleaning to remove any native oxide.

b- The Si wafers were moved to the sputtering chamber to Sputter a 1.4nm Al layer.

c- The Al layer was oxidized in situ (inside the chamber) for 8 hours under pressure of 100 mbar to create an AlOx layer.

d- A 24nm cobalt layer was sputtered on the top of the AlOx.

Fig. c8: I/V-characteristics and G/V-characteristics for Co/AlOx-tunneling contacts on highly doped silicon.

Fig. c9: I/V-characteristics for an ohmic contact on highly doped silicon. The I/V curve is completely linear.
At the end of the sputtering process the sample has the following vertical structure:

Si  200nm/ 1.4nm AlOx/ 24nm Co

A preliminary electrical characterization is always performed on the structures in order to check for a tunneling I-V-characteristic.

The current-voltage characteristics (I-V) of the device were measured using the same setup as for the (Ga,Mn)As tunneling structures and also the I/V-characteristics looked very similar. In order to investigate the interfaces between the AlOx and the Si, two samples have been sent to UOXF.DK for TEM characterization. Figs. c8 and c9 show sets of I/V-characteristics obtained for tunneling contacts and ohmic contacts respectively. The tunneling contacts show the expected non-linearity, while the ohmic contacts show ideal linear I/V curves and low contact resistances.

Fig. c10 shows the I/V characteristics of samples fabricated at UT for UWUERZ. The characteristics are similar to those of the samples fabricated in Wuerzburg, however, we expect that the samples from UT have a better homogeneity over the sample.

Investigation of tunnel barriers using TEM

In Oxford detailed investigations were done on the structural properties of tunnel barriers on silicon. Two techniques were used: Energy Dispersive X-ray Spectrometry (EDS) using the Oxford Instruments ISIS-LINK System and Electron Energy Loss Spectroscopy (EELS) using the Gatan Image Filter system. The two techniques have different advantages and disadvantages, depending upon the material system being studied. Both of these systems look at the energy imparted from the incident electron beam into the material, which excites the electrons in the material from the ground state to a higher state. EDS examines the energy of the X-rays emitted when the excited electron relaxes back into its ground state. This means that the detector is located close to the sample, as shown in Figure c11. EELS examines the energy lost by the beam after exciting electrons in the sample to higher energy levels.
Therefore, the detector is located below the sample, after an energy-dispersive prism (see Figure c13). By selecting a characteristic energy range, different elements and their location can be determined, both quantitatively and qualitatively. This is done using energy filtered TEM (EFTEM) for EELS or by elemental mapping for EDS. The advantages and disadvantages of EDS and EELS depend primarily upon what material system you are looking at and the conditions around the microscope on the day (i.e. how stable the sample is). EDS is a faster process, achieving better signal to noise ratios in a short period of time. EELS takes significantly longer – by a factor of 10 easily – to obtain the same SNR levels. This makes drift a major issue with EELS – too much drift and the signal smears out. Also, the background in EDX is essentially zero (see Figure c12). This makes it very easy to identify
the characteristic energy peaks. In order to detect the signal in EELS (see Figure c14), the curved background must be removed, adding another layer of difficulty. However, the energy resolution of EELS is significantly better than that of EDS. On the other hand, depending upon the elements in the sample, either EELS or EDS is better for a particular combination.
of elements. For example, for EDS, Si, Ta, and Zr are close together and therefore difficult to distinguish.

**Samples Investigated**

After the recent investigation of the electrical characteristics of Si$_3$N$_4$ and SiO$_2$ on Si, it was determined that either the barrier was not a good spin tunnel barrier due to hopping conduction (for Si$_3$N$_4$) or it was unstable electrically (for SiO$_2$). Therefore, recent work
focussed on transferring the existing technology of good spin tunnel barriers (Al$_2$O$_3$ and ZrO$_2$) onto Si. There were several objectives of this study – (1) to look at the diffusion of the metal and oxide into the silicon, and (2) to examine how the removal of the native oxide on the Si affected the underlying Si. It should be clarified that the samples all have the form shown in Figure c15 – the oxide tunnel barrier is sandwiched between the Si substrate on the bottom and a Ta capping layer on the top.
The first studies involved depositing $\text{Al}_2\text{O}_3$ and $\text{ZrO}_2$ directly onto Si (after removal of the native oxide by a smooth etch) with the same deposition conditions for the formation of high spin-conducting tunnel barriers on metals (oxide is formed by remote plasma oxidation). The results are shown in Figure c16 and Figure c17, where the left image (a) is always the HRTEM image and the right image (b) is either an EFTEM or EDS map of the same area. The $\text{Al}_2\text{O}_3$ sample showed significant diffusion of both the Al and the oxygen into the Si substrate. This has the potential to form an AlSi metal contact directly onto the Si semiconductor, destroying the spin polarization (in a worst case scenario) and in the best case, simply enlarging the potential barrier that must be crossed. On the other hand, the Zr and oxygen diffused very little, although the interface is not perfect nor completely uniform. In addition, the Si underneath the tunnel barriers, due to the diffusion, does not remain crystalline.

The next step involved depositing $\text{Al}_2\text{O}_3$ and $\text{ZrO}_2$ onto a thin SiO$_2$ layer, which was chemically formed by oxidation after removal of the native oxide. The deposition conditions were the same as for the above samples. The results are shown in Figure c18 and Figure c19, where the left image (a) is always the HRTEM image and the right image (b) is either an EFTEM or EDS map of the same area. The $\text{Al}_2\text{O}_3$ sample showed a very clean interface with no diffusion of either the Al or the oxygen into the Si substrate, at least on the resolution of the microscope. The $\text{ZrO}_2$ sample showed similar results to the previous sample, with very little diffusion of either the Zr or the oxygen. Again, however, the interface is not perfect. Finally, the Si underneath these samples was found to remain crystalline to within a couple atomic layers or less of the tunnel barrier.

**Electrical Characterization**

The promising results from the TEM work on the spin tunnel barrier $\text{ZrO}_2$ onto Si lead to the fabrication of 4-point probe devices to examine the IV characteristics of these tunnel barriers. These tunnel barriers were fabricated in exactly the same manner as the tunnel barriers discussed in the previous section. The only differences were the additional processing
to define the size of the contact to the Si and the deposition of Al on top of the Ta capping layer which was used to form the large-scale electrical contacts for wire bonding. A typical mask is shown in Figure c20, where the green lines define the Al electrical contacts and the small red box defines the size of the tunnel barrier on Si contact.

Two types of structures were fabricated: (a) a single-barrier structure where one contact to the Si uses the tunnel barrier and the other is a direct-ohmic contact to the Si and (b) a double-barrier structure where both of the contacts to the Si use the tunnel barrier. Furthermore, the tunnel barrier contact was varied in size between 1 mm x 1 mm and 1 mm x 20 mm to see if that would have any effect on the IV characteristics and the tunnel barrier parameters (average barrier height, average barrier thickness, etc.). (The same contact size per device was used for both of the Si contacts, regardless of whether it was a single-barrier or double-barrier device.)

Unfortunately, the initial measurements showed a surprisingly large resistance both with and without the presence of a tunnel barrier. This raises questions about the quality of the Si used, the existence of a good ohmic contact, or the formation of a Schottky barrier at the semiconductor-tunnel barrier interface. If the Si itself has too high of a resistance (or there is a Schottky barrier), then most of the voltage drop would be across the Si (or the Schottky barrier), and it would be difficult to determine any tunnel barrier characteristics due to the relatively small voltage the tunnel barriers would see. Alternatively, without the presence of
a good ohmic contact, quantitative data about the tunnel barriers cannot be determined.
Part of Oxford's third-year objectives is to heavily ion dope the top surface of a Si wafer in
order to ensure an ohmic contact between the metal and the Si and to reduce the presence
of any Schottky barrier (height and/or depletion width) and then repeat the electrical
characterization measurements.

In the mean time, although no quantitative data can be determined from the IV
characteristics, some overall "stability" characteristics can be observed:

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**Fig. c21**: Settling time of a tunnel barrier.

**Fig. c22**: Degradation of a Tunnel Barrier.
1. The IV Plot is sensitive to the amount of light; the sample resistance changes in the same manner as a solar cell. Furthermore, just shining a light on the sample will generate a non-zero voltage across the contact pads.

2. When voltage is applied to the devices, there is no breakdown up to 30 Volts (the maximum available voltage).

3. When current is applied to the devices, the breakdown occurs by approximately 0.5mA/mm².

The first major observation is that for similar currents, the measured/applied voltages are very different. This is a direct result of the respective manners in which the measurements were made. In the applied voltage measurement, the voltage is applied and the current is measured immediately. In the applied current measurements, the current is applied and the voltage is measured immediately. It is the immediateness of the measurement that causes the problem. Some of these barriers have extremely long settling times, as shown in Figure c21. Application of a current forces a particular current through the tunnel barrier, and it then takes a while for the charges to settle into the steady-state. Until that point, the measured voltage actually underestimates the final voltage. In contrast, the applied voltage forces a voltage drop across the constituent components, and the current has time to fluctuate therein until the charge distribution settles into the steady-state. This means, that the initial currents are higher than the final measured currents would be.

Again, it is quite unfortunate that the overall resistance of the system is so high. However, some general rules can be determined in regards to stability and current density that can be maintained. Specifically, for the thinnest barriers, they can maintain higher current densities for a given voltage, but they cannot hold it for very long (a few minutes or less) before degrading dramatically. The thicker tunnel barriers are more stable, and can maintain their current density over the barrier for longer periods without degrading. However, the required current density to yield the same voltage is less for the thicker barriers than for the thinner barriers.

The degradation of the tunnel barriers is quite surprising result itself. First of all, once the degradation begins, it acts like an avalanche – a large change results, which then briefly stabilizes before another large change happens, which repeats until the pinhole makes it all the way through the barrier and a short occurs. This is illustrated quite dramatically in Figure c22.

Spin injection into silicon (non local measurements)

UWUERZ has a long standing expertise in spin-transport experiments. From former experiments and also from experiments reported by other groups, we know that in a two terminal magnetoresistance measurements Hall effect and magnetoresistance in semiconductors can easily yield spin valve signals that can be misinterpreted as spin injection. Also the experiments on (Ga,Mn)As tunnel structures with non-magnetic metals indicate that spin valve signals can stem from unexpected and even unknown effects.

Because of this knowledge all transport experiments at UWUERZ are carried out in a four terminal and mostly also in a non-local geometry which is described in Fig. c23. The basic
principle of this geometry is that two contacts are used to induce a spin accumulation by injecting a spin polarized current. Two adjacent contacts which are placed outside the current path can be used to pick up the spin accumulation by selectively detecting the electrochemical potentials for spin-up and spin-down. In principle, the same measurements could be done with two pairs of a non magnetic and a magnetic contact, respectively, however due to technological reasons all contacts were fabricated as ferromagnetic contacts in a single step. The contacts are parallel lines with different width in order to allow for parallel or antiparallel alignment of the injection as well as of the detection contacts. Arrays of a large number of identical sets of contacts that correspond to the geometry used in Wuerzburg were sent to IPPAS. On these samples SQUID measurements are currently carried out in order to investigate the switching behaviour and the shape anisotropy. In a first step the the fingers were defined using e-beam lithography, evaporation of Ti/Au and lift off. Using this metallization as a mask the magnetic layer and the tunnel barrier
are patterned into fingers by Chemically assisted ion beam etching (Fig. c24a). On top of this layer negative lithography is performed leaving resist in the places where the contacts to the fingers will be made in a later step. A SiO layer is evaporated and lift-off is performed in such a way that now the whole sample except the contact windows are covered by an insulating layer (Fig. c24b). On top of this layers bond pads and the electrical interconnects to the fingers are deposited, leaving the sample ready for bonding (Fig. c24c).

Up to now the electrical measurements that need a magnetic field are performed in a room temperature magnet that was fabricated for this project. This magnet does only allow for very small magnetic fields. However they are sufficient for the measurements and the magnet allows for a sub Oersted field control.
Fig. c25 shows measurements that were performed in a non-local geometry. No spin injection could be detected up to now, however, the measurements already show that no side effects are visible in this geometry. The measurements show a lot of noise, which is probably due to insufficient shielding of the sample holder. A new sample holder is under construction. In the near future also measurements at low temperature will be performed. With regard to the typical performance of our measurement setups we hope that we can reduce the noise level to values close to the Johnson noise.

Fig. c26: Schematic of the fabricated spin transistor with silicon base.
Spin Injection into Si (vertical measurements)

The month 24 deliverable of ”spin injection into Si“ was heavily investigated at UOXF.DK. This was not performed on the standard tunnel barriers of $\text{Al}_2\text{O}_3$ and $\text{ZrO}_2$, but on another oxide tunnel barrier that is easier to fabricate on Si – namely $\text{Si}_3\text{N}_4$. ($\text{SiO}_2$ was also tried, but was found to be highly unstable.) In Oxford the samples were fabricated using standard photolithography on n- and p-type Silicon-On-Insulator (SOI) wafers. The final structure is shown in Figure c26.

Electrical characteristics in zero applied magnetic field

Two-terminal I-V characteristics

I-V characteristics of the emitter/collector (EC) junction and the emitter/base (EB) junction were performed at room temperature. As seen in Fig. c24 and c25, the results differ slightly between the two types of transistors, although the overall form is the same in both. Furthermore, the overall form is the same regardless of whether it is the emitter to base or the emitter to collector circuit. Since the emitter to base connection contains only one tunnel barrier (recall that the silicon to base junction is ohmic) and the emitter to collector contains two tunnel barriers, this implies that the emitter to silicon tunnel barrier dominates the electrical characteristics, yielding only non-linear tunneling curves.

For these $\text{Si}_3\text{N}_4$ barriers, the functional form of the I-V curves (including a linear resistance to account for parasitic currents through/around the barrier) close to zero bias is given by:

$$I = \text{I}_0 e^{B(V-V_0)} + CV + D$$  

Equation 1


For voltages greater than $\pm 0.5$ V, the form of the curves clearly changes and the functional form of hopping conduction no longer accurately fits the measured data. (The individual fits are shown in ??? and ???) This change is similar to that observed [C. Chaneliere, J. L. Autran, S. Four, et al., *J. Non-Cryst. Solids* **245**, 73 (1999)] in double structures of Al/$\text{Ta}_2\text{O}_5$/$\text{Si}_3\text{N}_4$/$\text{n-Si}$, which is ascribed to Fowler-Nordheim tunneling conduction [R. H. Fowler and L. W. Nordheim, *Proc. Royal Soc. London A*, **119** (1928) 173] overtaking the hopping conduction. The functional form for Fowler-Nordheim tunneling also accurately fits our data.
for voltages greater than $\sim |0.5|$ V, although the actual transition points between the regimes were determined by numerically differentiating the I-V curves.

$$I = AV^2e^{\frac{B}{V}} + CV + D$$

Equation 2

Moreover, the factor of four difference between the onsets ($\sim 0.5$V in our spin transistors compared to 2V in the double structures [C. Chaneliere, J. L. Autran, S. Four, et al., J. Non-Cryst. Solids 245, 73 (1999)]) correlates very satisfactorily with the thickness difference (2nm in our barriers as opposed to 8nm in the double structures). No systematic study of the onset of the change has been studied either by ourselves or in the literature; however, given the strong dependence of tunneling on the thickness of the barrier, this seems to be a reasonable hypothesis.

Fig. c27: Magnetic response of the spin transistors (a) before milling and (b) after milling.
There are two additional methods of confirmation possible. The first would be to observe the distinctive temperature dependence of hopping conduction \( I = I_0 e^{-\left(\frac{T_0}{T}\right)^{1/4}} \) [D.K. Paul and S.S. Mitra, Phys. Rev. Lett., 31 (1973) 1000] where \( T_0 \sim 7 \times 10^7 \) K. However, it was not measurable for these transistors due to carrier freeze out in the silicon at temperatures below 100K. The second comes from the well-established observation [S. S. Manoharan, D. Elefant, G. Reiss, et al., Appl. Phys. Lett. 72 (1998) 984] that hopping conduction destroys the spin-polarization of carriers. Therefore, no magnetic sensitivity should be observed in the hopping conduction regime. This is verified in a later section and lends additional confidence to our interpretation of the conduction methods.

**Electrical characteristics in an applied magnetic field**

The magnetic response of the Co layers in the spin transistors was measured using a vibrating sample magnetometer (VSM). The silicon background was subtracted from the measurement in order to yield Figure c27. These hysteresis loops indicate that differential switching is occurring in the devices. However, this experiment measured the switching fields of both the top and bottom magnetic materials simultaneously. By removing the collector and base contacts on the top surface (using ion milling with Argon ions – at 5keV, 0.8 mA, and an incident angle of 12 degrees – until the buried oxide layer was visible), further analysis indicates that the emitter (bottom) contact switches at the lower field of 40 Oe.

Application of a magnetic field is expected to affect the I-V characteristics in two ways. First, the magnetization of the emitter and collector Co contacts can be differentially manipulated, thereby introducing a spin-selective tunneling magnetoresistance (TMR) effect that modulates the collector current. Second, the applied magnetic field decreases the mean free path in the silicon base via Lorentz magnetoresistance (LMR) thereby also affecting the collector current.

**Two-terminal I-V characteristics**

The two-terminal measurements were repeated with a magnetic field applied in the plane of the transistor (perpendicular to the current). There are three important results (shown in Figure c28) from these measurements. First, the I-V characteristics are a function of applied magnetic field. Second, no magnetic sensitivity is observed for voltages below the onset of Fowler-Nordheim Tunneling (as indicated by the red arrows). This further substantiates the claim that hopping conduction is occurring at low voltages. Third, most of the activity (shown in the inset of Figure c28 as deviations from a straight line) in the electrical characteristics occurs around the magnetic transition region, between -90 and -115 Oe. These deviations (of magnitude >0.01 V) are outside of the noise level of the measurement, which is ±0.002 V. Furthermore, the fields at which the deviations occur stay consistent from measurement to measurement, although the exact voltage location and amplitude varies. This suggests that the deviations may be due to magnetic domain formation and/or motion in the Co layers (which are significantly larger than the single domain size of Co) changing the magnetic state seen in different regions of the tunnel barrier, since neither the deviations nor the magnetic domain pattern would be exactly reproducible. (Attempts to investigate the magnetodynamics...
by magnetic second harmonic generation or MFM were foiled by the thickness of the Al electrical capping layer, and the inability to remove it and retain a smooth surface.

Other proposed possibilities include fringe fields and anisotropic magnetoresistance. If the activity is the result of fringe fields, then the changing domain structure would generate stray fields which would change the local LMR as well as inducing a Hall effect, thereby changing the I-V characteristics. This is certainly possible and cannot be separated from the domain wall motion arguments at this stage. (If the electrical activity is due to domain walls, then the proposed next generation of transistors, which would be mono-domain, should not exhibit this effect.) AMR only occurs when the current flows parallel to the magnetization direction. In these devices, the current flow is always perpendicular to the magnetization direction, unless the magnetization rotates out of plane. In order for the magnetization to

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**Fig. c28: The two-terminal characteristics from Emitter to Collector as a function of applied**
rotate out of plane, the demagnetization field must be large compared with the anisotropy constant, which can occur in thin films. Assuming that the magnetization would rotate out of plane, AMR has only a 0.5% effect, normally. For the metal Co, which has a nominal resistivity of $6 \times 10^{-6} \, \Omega \cdot \text{cm}$, this translates into a resistance of 40 n$\Omega$. A percentage change of 0.5% of this is negligibly small (0.2 n$\Omega$), and therefore, does not account for the strength of the changes due to magnetic field.

LMR in the silicon is definitely measured (see Figure c31). (Any LMR in the ferromagnet, since it is only 30 nm thick and the silicon is 2 im thick, would be negligibly small.) This means that the Hall effect will also exist in the silicon, and will have a different sign depending upon what the sign of the carrier is. However, the Hall effect requires a measurement of the
voltage perpendicular to the direction of current flow. Measuring parallel to the direction of the current will just generate an increased resistance. Since the circuit only measures changes parallel to the current and not perpendicular, any hall voltages should not be picked up. However, if the fringe fields exist, it is possible that right at the edge of the tunnel barrier, the Hall effect could generate a spurious voltage. Finally, it has been observed previously that different domain structures form depending on the direction of magnetic field sweep – positive or negative [F.J. Jedema, M.S. Nijboer, A.T. Filip, and B.J. van Wees, cond-mat/0111092]. This causes the contacts themselves to have a MR behavior, which cannot be distinguished from the TMR effect. This is a valid point in this geometry, especially since the hypothesis is that the electrical activity is due to domain wall motion/formation. Therefore, only the known contributions can be removed – e.g. LMR, so the MR of the contacts could be dominating the MR signal.

Fig. c30: Collector current as a function of applied magnetic field of the spin transistors in common-emitter configuration with $I_B=0.6$ mA: (a) Transistor II-6 and (b) Transistor II-8.
Three-terminal I-V characteristics

The transistor was operated in common-emitter mode (sign conventions the same as for the NPN bipolar transistor) with the magnetic field applied in the plane of the transistor (perpendicular to the current). The results (Figure c29) show a variation in the collector current as a function of magnetic field. It should be noted that while the large deviations from a straight line occur in Figure c28, they do not appear in Figure c30. This is a direct result of the measurement in Figure c28 being two-terminal and in Figure c30 being three-terminal. The addition of the base current in the three-terminal configuration has a greater influence on the collected current due to its impact on the collector tunnel barrier height than the domain wall motion/formation and furthermore acts to stabilize the circuit as a whole.

The maximum variation of the current gain was -12 ± 4 % (-14 ± 3 %) for p-type (n-type) which occurred at -60 Oe and $I_B = 0.6 \, \mu A$. Overall, the current gain percentage shows a negative change – the collector current is being decreased as a function of field. The large oscillations in the current gain occur at the particular fields where there is anti-parallel alignment.

Fig. c31: Effect of an applied magnetic field on the collector current for Spin transistor II-6.
Micromagnetic measurements

For the investigation of spin transport in multy terminal devices (as in the non local geometry) it is important to know which relative magnetization the stripe like contacts exhibit at a certain point of the hysteresis loop. The magnetization is mainly determined by the shape anisotropy in such a way that the narrow stripes exhibit a higher switching field. For a detailed investigation of the magnetic properties, arrays of contacts with a layout similar to the structures that are used in the measurement have been fabricated by UWUERZ (Fig. c32). The structures were sent to IPPAS. Because of the large number of structures ($10^6$) the total magnetic moment is such that it can be detected using a SQUID magnetometer. In the magnetization measurements we should be able to identify the separate switching of the wide stripes and the narrow stripes, respectively. The magnetization measurements are currently under way.
Part 2/3: Work Progress Overview Year 3

Description of work and results

In the following four chapters the work of the partners on the different tasks is described in detail. According to the main efforts during year 3 we have dedicated the chapters to detection methods for spin injection (WP1), Schottky barriers and hot electrons (WP1), electrical measurements and spin injection into silicon (WP1), and Manganite based contacts (WP2) the latter describing the effort of growing high quality LSMO on semiconductor substrates and heterostructures and the characterization of the semiconductor structures after deposition and the electrical properties of the interface.
Part 2/3a (WP1): Novel detection methods

Introduction

The following part of the report deals with novel detection methods for spin injection into semiconductors. It reports results from measurements using the Oblique Hanle Effect (More measurements can also be found in the chapter on hot electrons and Schottky barriers). Also the improved results of the optical detection of spin accumulation by Faraday rotation and ultra fast optical spin injection and detection will be presented. In addition work on linear dichroism in (Ga,Mn)As is shown.

Time-Resolved Magneto-Optical Study of Epitaxial (Ga,Mn)As Films

The photo-induced spin polarization in epitaxial (Ga,Mn)As films was found to be equivalent to the application of an external magnetic field of about 1 mT for an optical excitation with a fluence of 10 µJ/cm². This photo-induced magnetization is one order of magnitude less than the one reported earlier [A. Oiwa et al, Phys. Rev Lett. 88, 137202 (2002).], and is found to relax with a decay time of 30 ps (Fig. 2), which is attributed to the spin relaxation of electrons in the conduction band. Raising the sample temperature above the Curie point, as well as application of a large inplane magnetic field does not result in any significant modification of this relaxation. From this we conclude that the spin relaxation of electrons is not much affected by Mn the ions. Therefore, spintronic devices based on (Ga,Mn)As can operate using only hole currents. Regarding the lower mobility of holes in comparison with that of electrons, this condition will limit the operation speed of the (Ga,Mn)As-based devices.

Figure 1. Dynamics of the Kerr rotation recorded at 10 K, compared with similar data from the reference sample containing a LT-GaAs film. The photon energy was 1.56 eV. The solid lines are fits to the experimental data.
Optical Hot Electron and Spin Injection in InP:

The knowledge of the spin lifetime in semiconductors is an important parameter, which is necessary for the design of the novel spintronics devices. Beyond the gigahertz frequency domain, the spin dynamics can only be studied by optical techniques, such as polarized photoluminescence or magneto-optical effects. Both magneto-optical Faraday effect in the transmission geometry and Kerr effect in the reflection geometry are well suited for studies of the spin dynamics in a broad spectral – including wavelengths where no luminescence can be measured - as well as thermal range.

The goal of the following study was the injection of hot spin-polarized electrons and holes into InP semiconductor with subsequent observation and understanding of picosecond and sub-picosecond dynamics of electron and hole spins in intrinsic and p-doped InP in the broad temperature range from 20 K up to 300 K.

The study was done using the time-resolved, pump and probe, photo-induced magneto-optical Kerr effect. For the experiments a pulsed Ti:sapphire laser was used, with a pulse duration of approximately 100 fs and a repetition rate of 82 MHz. The measurements were done in the spectral range from 1.47 eV to 1.57 eV nearby the transition between the split-off valence band and the conduction band (E_g =1.53 eV). The Kerr rotation and ellipticity
experienced by the reflected delayed probe pulses were measured as a function of the time delay between pump and probe pulses.

![Graph showing MOKE rotation and ellipticity as functions of time delay and photon energy.](image)

**Figure 3.** Time dependence of the MOKE rotation induced in InP sample measured with pump-probe technique.

The typical temporal behavior of the photoinduced Kerr rotation is shown in Figure 3. Three relaxing components with decay times of about 10 ps, 20 ps and more than 300 ps can be distinguished in the magneto-optical signal. The spectral behaviour of these components was analysed. On the base of the obtained spectra, theoretical predictions and simulations the component with decay time 10 ps (Fig. 4) was shown to be not relevant to spin dependent phenomena but rather caused by phase space filling effects.

![Spectral dependence of three different relaxation processes.](image)

**Figure 4.** Spectral dependence of three different relaxation processes characterized by relaxation times of 10 ps (a), 20 ps (b), and 300 ps (c). Both MOKE rotation and ellipticity are plotted.
We have also analyzed theoretically the spectra of the magneto-optical response for highly nonequilibrium spin distribution in semiconductor InP. A specific contribution to the photo-induced magneto-optical Kerr effect is distinguished in the spectral region where the transitions from spin-orbit split-off valence band to conduction band are excited. This contribution is characterized by a fast $\ll 1$ ps relaxation time (see Fig. 5) and interpreted to be due to a partial compensation of spins excited from the heavy-hole, light-hole and spin-orbit split-off valence bands in the process of electron thermalization.

![Figure 5](image1.png)

Figure 5. Temporal behavior of the Kerr rotation (a) and ellipticity (b) at different photon energies and $T=20$ K. One can see the almost instantaneous drop of the Kerr signal at the time scale of about 1 ps.

![Figure 6](image2.png)

Figure 6. The calculated electron charge $n(E)$ (solid line) and spin $s(E)$ (dashed line) distributions in the conduction band InP created by a pump pulse in the absence of population relaxation at 1.55 eV (a) and 1.7 eV (b). The rapid variation of $n(E)$ and $s(E)$ at small energies at 1.55 eV is due to the Coulomb enhancement factor. The quasiequilibrium electron spin distribution corresponding to the effective electron temperature of 600 K is shown by dotted line for comparison. (c) The calculated dependence of the Kerr rotation and ellipticity on the photon energy in InP.
Fig. 6 shows the calculated charge and spin distribution of photoexcited hot electrons. Three bands of the distribution corresponding to excitations from HH, LH and SO valence bands are clearly seen. A peculiar property of the photoexcited population is that the spin distribution changes sign so that total electron spin is reduced due to a partial compensation of spins excited from different valence bands. At photon energies slightly above the SO band gap the spin distribution $S_{so}$ of electrons excited from the SO valence band has opposite sign with respect to $S_{lh}$ and $S_{hh}$. When the photon energy increases, $S_{so}$ tends to zero but $S_{lh}$ and $S_{hh}$ becomes equal in magnitude and opposite in sign so that the total spin density of the hot electrons approaches zero.

**Ultrasensitive Magneto-Optical Detection of Spins Electrically Injected Into Bulk Semiconductor**

Most studies of the optical studies of spin injection into semiconductors so far were based on the measurements of photoluminescence, whereas this method has considerable limitation on sample, spectral and thermal range. Moreover the study of electrically injected spins based on the measurements of photoluminescence can not supply quantitative estimate of the number of spins injected. The magneto-optical Kerr effect provides a good alternative to the well-known photoluminescence technique. This effect can be adopted for the study of spin injection with no limitation with respect to sample as well as in broad spectral and thermal range including wavelengths were no luminescence can be measured. Here on the example of the heterostructure ZnMnSe/GaAs we demonstrate that the magneto-optical Kerr effect is a powerful tool for the study of electrical spin injection.

For the measurements we used ultrasensitive laser polarimeter. The light was directed from the side of ZnMnSe. After transmission though this spin injector the light was reflected from the interface ZnMnSe/GaAs. The wavelength of light was chosen to be 810 nm so that ZnMnSe is transparent at this wavelength, but absorbtion in GaAs is quite large and the reflection at the interface ZnMnSe/GaAs is effective (see inset in Fig. 7). Moreover, the energy of photons at this wavelength is close exciton resonance in GaAs and all magneto-optical effects in this semiconductor should be resonancely enhanced. The sample was placed in the static magnetic field directed along the normal of the sample so that spins in ZnMnSe were aligned in the direction of the magnetic field. We have sent rectangular pulses of current at frequency $\omega$ through the structure and detected the rotation of the polarization of the reflected light at this frequency.

The magneto-optical Kerr effect induced by spin polarized current in ZnMnSe/GaAs heterostructure is shown in Fig. 7 as a function of an external magnetic field. It is seen that the effect depends linearly on the external magnetic field. Such linearity originates from the fact that the spin injector ZnMnSe is paramagnetic material. For magnetic field used in our experiment paramagnetic susceptibility is far from saturation and efficiency of spin injection should be proportional to the external magnetic field.

Fig. 8 shows the measured magneto-optical signal as a function of the voltage applied to the structure. One can see that magneto-optical signal is not detected if the voltage is positive and current is directed from GaAs into ZnMnSe. However, if the voltage is negative and current flows from ZnMnSe into GaAs a pronounceable magneto-optical signal is
detected. The voltage dependence of the magneto-optical signal is characterized by the well distinguished maximum at 0.7 V. Inset in Fig. 8 shows IV-characteristic of the studied structure.

Figure 7: The magneto-optical Kerr effect on the frequency of the applied voltage as a function of the external magnetic field. The dependence is obtained at photon energy 1.52 eV. The voltage was applied in form of the rectangular pulses of negative polarity. The amplitude of the pulses was equal to 1.2 V. The inset shows the geometry of the experiment.

Figure 8. The magneto-optical Kerr effect on the frequency of the applied voltage as a function of the amplitude of the pulses. The dependence is obtained at photon energy 1.52 eV with an external magnetic field equal to 0.3 T.

**Giant Magnetic Linear Dichroism as a Key to the Electronic Structure of Ferromagnetic (Ga,Mn)As**

One of the most popular optical methods to study and demonstrate the magnetic properties of zinc-blende semiconductors is based on the measurements of their photoluminescence, as
recombination of spin-polarized carriers leads to circular polarized radiation. However this method appears to be impractical for (Ga,Mn)As as it hardly shows any detectable photoluminescence. This is most likely due to the large number of defects which appear because of the low-temperature growth conditions and act as nonradiative decay centers. Another frequently used technique to investigate the magnetic properties of media is based on measurements of magneto-optical phenomena such as Magnetic Circular Dichroism (MCD) and Faraday or Kerr effects, that are of first-order in the magnetization. There were several attempts to apply these techniques for the study of the electronic structure of (Ga,Mn)As, but the interpretation of the magneto-optical spectra of (Ga,Mn)As appeared to be a complicated problem. Moreover, the sp-d interaction in Mn-doped semiconductors is responsible for a strong magneto-optical signal already in the paramagnetic phase, so that the ferromagnetic order does introduce only small changes in the spectra. Thus it is clear that for the progress of understanding the electronic and magnetic properties of these novel ferromagnetic semiconductors, alternative techniques are required.

We make here a breakthrough by observing a giant Magnetic Linear Dichroism (MLD) in (Ga,Mn)As, which is about two orders of magnitude larger than that reported for this material earlier. We show that, in contrast to MCD, this effect is predominantly sensitive to the specific electronic states in the valence band, which are responsible for the hole-mediated ferromagnetism in (Ga,Mn)As. This makes MLD to be a powerful tool to study the electronic structure of ferromagnetic semiconductor alloys such as (Ga,Mn)As.

The field dependence of the MLD is characterized by an M-shaped hysteresis loop with jump-like changes of the signal at $H_{12} = 9 \text{ mT}$ and $H_{23} = 110 \text{ mT}$. Such hysteresis form is typical for the magneto-resistance in (Ga,Mn)As as well as for other effects which are even with respect to an external magnetic field or magnetization. This M-shaped hysteresis loop for (Ga,Mn)As originates from the four-fold magnetic anisotropy of this material. The process of magnetization reversal in an external magnetic field happens via jumps of the magnetization over $90^\circ$, so that in a relatively weak field, the magnetization is practically parallel to one of the [100] or [010] easy axes.

In order to demonstrate the selective sensitivity of the MLD to ferromagnetism induced changes in the electronic structure of (Ga,Mn)As, we compared the MLD spectrum with that of the magneto-optical Kerr effect (Fig. 10(a)). In the studied spectral range the Kerr effect exhibits two peaks at 1.6 eV and 1.9 eV, respectively. It was shown that the low-energy peak around 1.6 eV shifts with the Fermi energy when the hole density is varied. Therefore the 1.6 eV peak is related to the interband transitions from the Fermi level, located inside the valence band, to the conduction band. This conclusion is in good agreement with the absorption spectrum shown in Fig. 10(b).

In contrast to the magneto-optical Kerr effect, that shows substantial amplitude in a broad spectral range, the linear magnetic dichroism exhibits a resonance only around 1.6-eV and stays almost zero for higher photon energies. This implies that transitions from states in the vicinity of the Fermi level are almost exclusively responsible for the giant MLD signal in (Ga,Mn)As.
Figure 9. (a) Sample orientation with respect to the applied magnetic field and the four-step magnetization reversal process; (b) Field dependencies of the magnetic linear dichroism (the MLD hysteresis loops) for different angles $\theta$ between the incident polarization and the [100] crystallographic direction in (Ga,Mn)As (indicated in the figure), measured at a wavelength of $\lambda=815$ nm.

Figure 10. (a) Polar magneto-optical Kerr effect and magnetic linear dichroism in the Ga$_{0.98}$Mn$_{0.02}$As film as a function of photon energy. (b) Absorption in Ga$_{0.98}$Mn$_{0.02}$As as a function of photon energy.
Magnetization switching in GaMnAs by femtosecond optical excitation

We demonstrate complete reversal of a full magnetic hysteresis loop of the magnetic semiconductor (Ga,Mn)As by ultrashort optical excitation with a single 100-femtosecond light pulse, with obvious implications for ultrafast magneto-optical recording. Our approach utilizes the four-fold magnetic anisotropy of (Ga,Mn)As, in combination with the linear magnetic dichroism of the material (see previous paragraph).

Figure 11. Hysteresis loops of the Ga$_{0.98}$Mn$_{0.02}$As sample with in-plane easy axes along the [100] and [010] crystallographic directions. The applied in-plane magnetic field makes an angle of 41° with the [100] direction. (a) The major hysteresis loop addresses all four possible magnetization states; (b) Minor hysteresis loops address only two states, lower 1 and 2 or upper 3 and 4. (c) Experimental major hysteresis loop measured via the linear magnetic dichroism. Dark and light colors correspond to up-sweep and down-sweep of the hysteresis loops. (d) Experimental lower minor hysteresis loop measured via the linear magnetic dichroism. (e) Schematic behavior of the magnetic birefringence signal accompanying a switch from state 2 to state 3 in a constant magnetic field at low temperature (bottom panel), when reducing the coercivity through a laser heat pulse (middle panel), and when returning to low temperature (top panel).

The ferromagnetic materials that are commonly used in magneto-optical memory devices possess uniaxial magnetic anisotropy, i.e. there is an easy axis, along which the magnetization can be either parallel or antiparallel. As a result, the magnetization as a function of external
magnetic field exhibits a hysteresis loop resulting from the switching between two metastable magnetization states. In contrast, the ferromagnetic semiconductor (Ga,Mn)As has a four-fold magnetic anisotropy, and as a result possesses two equivalent easy axes. This property modifies the hysteresis loop such that a switching of the magnetization between two different pairs of states can be observed. Here we demonstrate that this switching may be induced by applying a single laser pulse that can be as short as 100 fs. Moreover, each pair of the four magnetization states in the (Ga,Mn)As layer can function as a bit, leading to a doubling of the recording density. For the read-out of such memory states, magnetic linear dichroism can be used.

Figure 12. Minor hysteresis loops of the Ga$_{0.98}$Mn$_{0.02}$As sample, recorded before (light curve) and after (dark curve) single optical pulse with a duration of 100 fs, are shown together with corresponding magnetization orientations (insets).

**Magneto-optical diffraction as a tool to study the magnetization of sub-micrometer injecting contacts**

For the measurements, a pulsed laser beam from a Ti–sapphire laser (76 MHz ‘100 fs pulses) with a wavelength of 760 or 810 nm was focused onto the sample. The polarization of the incoming fundamental laser beam could be chosen using a Babinet Soleil compensator and a polarizer was used afterwards to ensure a high degree of light polarization. An analyzer was set to choose the polarization of the outgoing SHG, that was detected after proper filtering with a photomultiplier. For the MOKE hysteresis measurements, a Wollaston prism in combination with a differential 2-diode detection scheme was employed. The angle of incidence was always 45°, while the detection arm could be rotated in order to measure the magneto-optical signal as a function of the diffraction angle. The transversal magneto-optical geometry was used for most of the experiments. The samples, sandwiched Pt/CoNi/Pt films, were sputter-deposited and structured at University of Twente.

It has been shown experimentally that magneto-optic Kerr effect (MOKE) hysteresis loops recorded for diffracted beams are quite different from those recorded from the reflected beam (see Fig. 13). It was found that changes in shape and coercive field depend on diffraction order and easy axis direction. A rigorous theoretical solution to the diffracted Magneto-optic Kerr