1 Title of the project

Control of magnetism and conductivity in EuO

2 Applicant

M.C. Donker

Selected works:

- Ultrafast carrier and magnetization dynamics in EuO
  Master thesis, University of Groningen (2006)

- Structural, magnetic and transport properties of Ca-doped $PrVO_3$
  Bachelor thesis, University of Groningen (2004)

3 FOM- research group

GM-G-

4 Institute

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5 Abstract

In the field of spintronics one is looking for materials from which spins can be efficiently injected into a semicondutor. Because of the high degree of spin-polarization in the conduction band and the lack of a conductivity mismatch, the ferromagnetic semiconductor EuO is a promising spintronics material. EuO can be used as a ferromagnetic contact but also as a spin-filter. The proposed research is about the manipulation and control of the magnetism and conductivity in EuO films. This will be done chemically by doping, optically by laser pulses, electrically by applying a gate voltage in a EuO metal-oxide-semiconductor field effect transistor or magnetically by applying a magnetic field. By using combined optics and spin-polarized transport techniques, spin-filtering phenomena can be verified.
We will study the mechansim and speed of the magnetic and conductivity switching and look for the possiblity of making thermal and non-thermal phase transitions (i.e. ferromagnetic/paramagnetic and metal/insulator transitions) by ultrashort laser pulses. This will be done using time-resolved spectroscopy techniques. By optical techniques also spin-dephasing times, spin-mobilty and spin-lifetimes will be determined.

6 Duration of the project

4 years, starting from November 2006.

7 Personnel

7.1 Senior- scientists

Name	Main task	Time
Prof. dr. ir. P.H.M. van Loosdrecht	Management and supervision	10%

7.2 Junior-scientist and technician

Name	Main task	Time
M.C. Donker	Experiments and analyses	90%
Technician	Technical support	10%

8 Cost estimates

8.1 Personnel positions

One A.I.O. position for four years.

8.2 Running budget

15 k€/ year

8.3 Equipment

All the needed equipment is present.

8.4 Other support

The project will be supported by the Material Science Centre^{plus}. They will contribute in the positions of the technician and the senior scientist. Samples can be grown on a Molecular Beam Epitaxy (MBE) setup at the University of Cologne in the group of prof. dr. L.H. Tjeng at the Institute of physics II.

8.5 Budget summary in k€

	2006	2007	2008	2009	2010	TOTAL
personal (positions):						
PhD students	.3	1	1	1	.7	4
postdocs	-	-	-	-	-	-
technicians	-	-	-	-	-	-
guests	-	-	-	-	-	-
personal (costs)	12.9	43	43	43	30.1	172
running budget	4.5	15	15	15	10.5	60
equipment FOM-part	-	-	-	-	-	-
TOTAL (requested from FOM)	17.4	58	58	58	40.6	232

9 Research programme

A) Introduction
B) Goals
C) Description of the work
D) Plan of work

A. Introduction

Until recently the spin of the electron was ignored in electronic devices. However, apart from using the charge of an electron, the spin degree of freedom can be used either which offers opportunities for spin-electronic devices. The injection and detection of a spin-polarized current in a semiconductor could combine magnetic storage of information with electronic read-out in a single semiconductor device. To create this kind of devices, first one have to be able to generate a spin-polarized current in a semiconductor. Here fore several strategies has been used. The most simple one involves a ferromagnetic metal, like nickel, in contact with a non-magnetic semiconductor. By applying a voltage, one can generate a spin-polarized current in the semiconductor. Schmidt *et al.*[1] revealed that the basic obstacle for spin injection from a ferromagnetic metal into a semiconductor originates from the conductivity mismatch: the spin injection coefficient (γ) in the diffusive regime is $\gamma \sim \sigma_{SC}/\sigma_{FM}$ in which σ_{SC} and σ_{FM} are the conductivity of semiconductor and the ferromagnetic metal, respectively[2]. One can see that the spin injection coefficient is limited by the difference in the conductivity in the semiconductor to the conductivity in the metal. It has already been shown that this problem can be partly solved by using tunnel barriers for conductivity matching[3] but due to the limited spin-polarization in the ferromagnetic metal, the degree of spin-polarization of the current in the semiconductor will never reach 100 %.

Good candidates for spin-injection contacts are the class of ferromagnetic magnetic semiconductors. In these materials the conductivity mismatch between the magnetic and non-magnetic semiconductor can be very small and the spin-polarization of the conduction band electrons in a ferromagnetic semiconductor can be as high as 100%. This makes ferromagnetic semiconductors an interesting class of materials.

Apart from choosing a good material for spin-injection, one would like to be able to manipulate the magnetic and transport properties of the material in an electronic device. It should be convenient for instance when it is possible to change the resistivity by applying a magnetic field as can be done in a Colossal Magneto-Resistance (CMR) material, by applying a gate voltage as is done in Metal Oxide Semiconductor Field Effect Transistors (MOSFET's) or optically by introducing carriers in the conduction band. It should be convenient as well when the magnetic properties can be controlled in the same way, by an applied magnetic field, by a field-effect or optically.

Crucial in the control of the conductivity and the magnetism in a material is not only the ability to switch but also the switching speed (i.e. switching between good or bad conducting). In today's electronics this switching speed is usually relatively slow. In minidisks for instance, the magnetization orientation in a magnetic bit can have two directions. In order to flip the magnetization, the ferromagnetic material is heated above the ordering temperature by a continuous wave laser while at the same moment a magnetic field is applied. When the laser is turned off, the material can cool down and the magnetization will grow in the direction of the applied magnetic field. In this case the limitation for the writing speed is the thermal cooling, which can take nanoseconds. With the availability of femtosecond lasers, there came the idea to optically control materials in a non-thermal way in which the writing speed is not limited by the laws of thermal statistics and can be much faster. Numerous studies have been shown that it is indeed possible to induce non-thermal ultrafast phase transitions between for instance solid and liquid phases[4] or metallic and insulating phases[5]. These ultrafast switching processes are from technological as well as from fundamental interest as they provide a tool for making faster electronics but also give access to non-thermal states.

B. Goals

The proposed research involves a study towards the (ultrafast) manipulation of the magnetism and conductivity in an interesting spintronic material, namely the ferromagnetic semiconductor EuO, and a study towards the feasibility of using this material in spintronic applications. EuO (Eu-rich) has a Colossal Magneto Resistance (CMR) effect and has a fully spin-polarized conduction band[6]. The magnetic ordering temperature is dependent on the number of electrons in the conduction band. By doping EuO with Gd, the conduction band can be populated and the ordering temperature can be increased from 69 K (EuO) to 180 K (4% Gd-doped EuO)[7].

The goal of the research is to study the (ultrafast) magnetization and carrier dynamics, to understand the mechanism of (ultrafast) switching and finally to control the magnetism and conductivity in EuO. We will not only try to change the magnitude of the magnetization and the conductivity but also try to make the (ultrafast) ferromagnetic/ paramagnetic and insulator/ metal phase transition. The physical properties will be manipulated chemically by doping, optically by laser pulses, electrically by applying a gate voltage in a EuO metal-oxide-semiconductor field effect transistor or magnetically by applying a magnetic field.

Since EuO is an potentially interesting spintronics material, the spin-dephasing time, the spin-mobilty and the spin-lifetime will be determined in the paramagnetic phase. However, the first goal will be to make high quality stoichiometric EuO, Eu-rich EuO and Gd-doped EuO thin films on a MBE setup at the University of Cologne. The EuO MOSFET can be made on the same setup.

C. Description of the project

EuO and the other europium chalcogenides (EuS, EuSe and EuTe) have been studied in quite some detail over the last 50 years. Whereas EuO is ferromagnetic with a T_c of 69 K, EuS is ferromagnetic with a T_c of 16 K, EuSe is both ferro- and antiferromagnetic (T_N=4.6 K, T_c=2.8 K) and EuTe is antiferromagnetic with a T_N of 10 K. EuO is a semiconductor in which the magnetic moment reside on the europium ion ($4f^7$) [8]. The valance band consist of 4f-orbitals whereas the conduction band consist of hybridized oxygen 2p- europium 5d orbitals. EuO is ferromagnetic mainly because of an indirect exchange interaction mechanism in which an 4f electron makes a virtual transition to the conduction band where it experiences an exchange interaction with nearest neighbour europium 4f electrons and then returns to the ground state.

The exchange interaction in EuO is thought to be mediated by the conduction electrons. When EuO is doped with Gd, the divalent Eu ion is replaced by the trivalent Gd ion and the conduction band is populated. Tjeng and co-workers have shown that in this way it is possible to increase the ordering temperature of EuO by 110 K to 180 K in 4% Gd-doped EuO films[7]. Optimizing the doping concentration or the dopant may lead to even higher ordering temperatures. Apart from chemical doping we will also try to electrically dope EuO in a MOSFET structure. An EuO MOSFET structure is shown in figure 1 which consist of a substrate, an EuO film, a source (S) and drain (D), an insulator and a transparent gate electrode. By applying a gate voltage, a conductivity channel can be created. The conduction electrons in this channel may enhance the ferromagnetic exchange interaction resulting in an enhanced ordering temperature. Since Eu-rich EuO has an CMR effect, the transport through the semiconductor in the FET can be changed by an applied magnetic field. The magnetic properties of EuO in the conduction channel can be probed by the magneto-optical Kerr effect using linearly polarized light. The magneto-optical Kerr effect is the rotation of polarization upon reflection from a magnetic medium. The Kerr rotation is proportional to the magnetization and is very large in EuO, up to 7 degrees. Therefore it is an excellent tool for probing the magnetic properties in the conduction channel.

Another important property of EuO is that is has a spin-polarized conduction band. At 10 K the splitting between the spin-up and spin-down subbands is as high as 0.6 eV. This large splitting makes EuO a interesting spintronic material. EuO can be used as a ferromagnetic contact for spin injection or as a spinfilter. Moodera and co-workers have shown that the spin polarization in metal/EuO/metal tunnel junctions can be 29% at 0.1 T and 0.45 K [9]. Better quality EuO at the interface may increase the spin polarization towards 100 %, at higher temperatures and without applied magnetic field.

The temperature dependence of the splitting of the conduction band behaves like the magnetization. It is large at low temperatures and disappears at the magnetic ordering temperature [6]. The existence of the spin splitting has great consequences for the transport properties in Eu-rich EuO. In Eu-rich EuO there are oxygen vacancies. These oxygen vacancies can bind two electrons and the vacancy levels are situated in the bandgap. When by decreasing the temperature the ordering temperature is crossed, a insulator to metal transition is observed. It is thought that this is because of the crossing of the spin-up conduction band with the vacancy levels. Gd-doped has a MIT as well but no MIT is observed in stoichiometric EuO. High europium-oxygen non-stoichiometry can also lead to an enhanced T_c of 150 K [10].

When stoichiometric EuO is illuminated with 632.8 nm (> band gap) laser light, an MIT is observed as well. Above T_c illumination just leads to a lower conductivity but at T_c illumination leads to a change in the slope in the temperature dependence of the resistivity which is positive at below T_c and negative above T_c [6]. Steeneken proposed for this MIT a mechanism in which electrons in (magnetic) excitons becomes dissociated upon magnetic ordering and can contribute to the conduction. Apart from changes in the free electron concentration also changes in the mobility are important as was shown by Kajita and Masumi [11]. The mobility close to T_c (60 K) is dependent on the applied magnetic field and can be increased by 78 % when an magnetic field of 15 kOe is applied. We want to study the photo-conductivity in detail. For this we have a combined optics/transport setup which consist of an optical cryostat with superconducting magnet in which we can measure the conductivity. For these measurements, EuO films with contacts can be grown on a MBE setup, as is shown in figure 1. To get a better understanding of the MIT in the photoconductivity, the photoconductivity will be measured at different temperatures and magnetic fields and using different wavelengths and intensities.

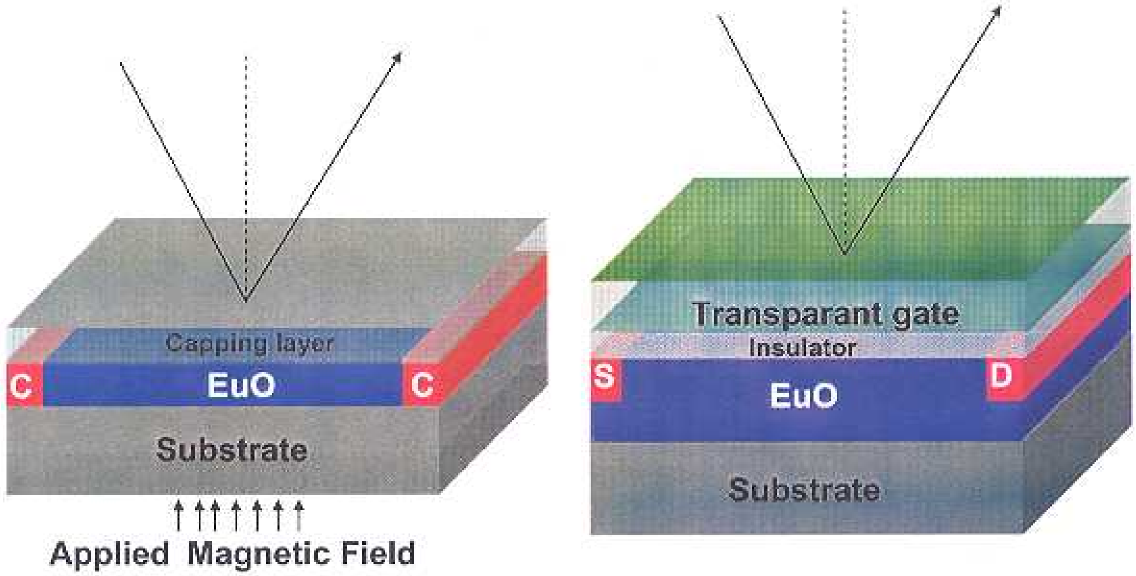

Figure 1. *In the left picture an (stoichiometric, Eu-rich or Gd-doped) EuO sample is shown including a substrate, EuO film, contacts (C) and a protective capping layer. The arrow indicates the incoming light. In the right picture an EuO MOSFET is shown. S and D stands for the source and the drain, respectively, of the MOSFET. The gate can be made of a transparent metal. In this way the conduction channel can be probed.*

Both the conductivity in Eu-rich and Gd-doped EuO are sensitive to an applied magnetic field. In Eu-rich EuO a large negative CMR of six orders of magnitude at T_c was observed. The CMR is described in a similar way as the MIT is described. An applied magnetic field can align the magnetic moments and because of this it can induce a spin-splitting of the conduction band. When the conduction band crosses a vacancy level in Eu-rich EuO, the electrons can dissociate and contribute to the conduction [6].

Apart from studying the influence of electrical doping in a MOSFET and chemical doping we are also interested in the ultrafast magnetization and carrier dynamics in EuO. It has already been shown in that in diluted ferromagnetic semiconductors, namely (Ga,Mn)As or (In,Mn)As, the ferromagnetism can be quenched at short timescales (< 1ps) using ultrashort intense linearly polarized laser pulses[12]. However it is also possible to increase the magnetization, using circularly polarized laser pulses[13]. This is thought to be because of the fact that circularly polarized photons can transfer angular momentum to the electrons when an electron make a transition from the valance to the conduction band. The photoinduced spin polarization was very small and was found to be equivalent to the application of an external magnetic field of about 1 mT.

Preliminary results of us on EuO show that is it possible to both increase and decrease the magnetization using linearly polarized laser pulses. The measurements were done on a non-amplified laser system providing 50 fs laser pulses with a central wavelength of 800 nm (1.55 eV) which is larger then the bandgap (1.2 eV). First a linearly polarized strong laser pulse, the pump pulse (1.55 eV), hits the sample after which a weaker linearly polarized laser pulse, the probe pulse (1.55eV), hits the sample on the same spot. By varying the delay between the two pulses and looking to the rotation of polarization (Kerr effect) of the reflected probe pulse, the magnetization dynamics can be studied. The used pump fluence is about 1 mJ/cm^2. By using this value the photodoping can be calculated which equals 0.4 %. In figure 2 a transient Kerr rotation hysteresis loop ($\Delta\theta$) is shown for -1 ps delay (probe arrives before pump) and for +1 ps delay (pump arrives before probe). A transient Kerr rotation hysteresis loop ($\Delta\theta$) is the difference between the normal hysteresis loop (θ) and the hysteresis loop with the influence of the pump pulse ($\theta+\Delta\theta$). The sign of the saturation magnetization (saturation Kerr rotation) at -1 ps is different from the sign +1 ps. By looking to the sign one can see that the hysteresis loop at -1 ps corresponds to an induced demagnetization whereas the hysteresis loop at +1 ps corresponds to an induced demagnetization. This means that because of the pump, the total magnetization can be increased or decreased. The signal at negative delay can explained by overall heating. When electrons are excited higher in the conduction band, they can relax by emitting phonons. Together with the heating from non-radiative recombination, this can lead to a heating of the sample. The repetition rate of the laser is 800 kHz which means there is one pump pulse per 1250 ns.

The increased magnetization (0.6%) at +1 ps is thought to be because of the increased population of electrons in the conduction which can enhance the exchange interaction. When the signal at -1 ps is subtracted from the signal at +1 ps (which is because of heating), the electronic effect is obtained which is shown in figure 2. Since this value is positive in sign, one can conclude that the electronic effect corresponds to an induced increase in the magnetization. The magnitude of the electronic effect is decreasing with temperature.

When transient hysteresis loops are measured at different delays, we can study the magnetization dynamics and monitor the remanence, the saturation magnetization and the coercivity on the same time. We can however also put a constant magnetic field, for instance +100 mT (which is higher than the value of the coercivity in EuO) and vary the delay. In this way the saturation Kerr rotation can be followed in time and we can study the electronic and the heating effects.

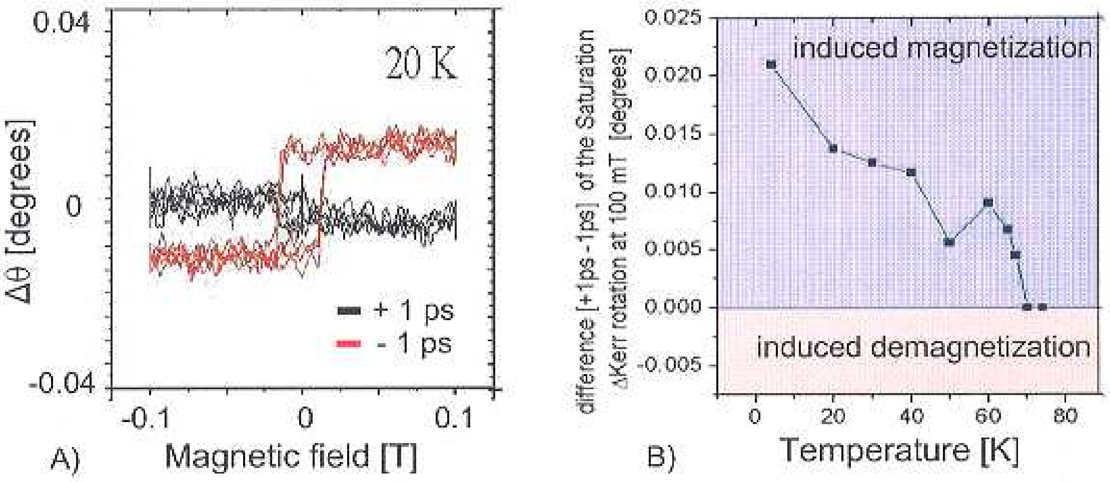

Figure 2. *A) Transient hysteresis ($\Delta\theta$) at 20 K for -1 ps and +1 ps delays. Both pump and probe pulses were 800 nm and linearly polarized. The pump fluence was about 1 mJ/cm^2 which corresponds a photodoping of 0.4%. B) Electronic effect, which is the difference in the saturation $\Delta\theta$ at +1 ps compared to -1 ps. When this signal is positive, it means that the electronic effect corresponds to an induced increase in the magnetization (Donker et al. , to be published).*

The induced changes in the magnetization are rather small, 0.6%. One thing that we can do to improve this is to repeat this experiment on an amplified laser system. Because of the increased pump fluence, the induced changes will be bigger and we will try to make a phase transition. We do an experiment at 72 K which is just above T_c in stoichiometric EuO and try to make a paramagnetic to ferromagnetic phase transition. The pump pulse can excite electrons to the conduction band. Since conduction electrons can enhance the exchange interaction, it might be possible to increase T_c to above 72 K and induce the phase transition. To minimize heating effects one can excite with 1.2 eV pump pulses. Since the electrons do not have to relax in the conduction band, heating effects can be reduced.

We can also go to 65 K and excite high in the conduction band and try to induce an ferromagnetic to paramagnetic transition by laser heating. By using different pump powers, pump wavelengths, pump polarizations (linear or circular), different applied magnetic fields and different Gd concentrations we can study the mechanisms of the optically induced effects and characterize (e.g. speed) the phase transition. As discussed, apart from chemical doping we can also electrically dope EuO. By using an transparent gate electrode in the MOSFET, it is possible to probe the ultrafast magnetization dynamics in the conduction channel which can give additional information. For a full characterization of the induced effects the carrier dynamics have to be studied as well. On our setup, the magnetization dynamics and the carrier dynamics can be studied on the same time. This can be done by measuring changes in the reflectivity, which is sensitive to the number of carriers in the conduction band. Time-resolved photoluminescence experiments can be done complementary.

Apart from trying to make a ferromagnetic-paramagnetic phase transition we can also try to make an insulator-metal transition. By applying an magnetic field to an Eu-rich EuO sample, the CMR effect can be manipulated optically by strong pump pulses and probably an insulator to metal transition can be induced. The transport properties can be probed using time-resolved THz-spectroscopy which is sensitive to the number of free electrons and is a way to measure the transient photoconductivity. Another way in which the conductivity in Eu-rich EuO can be manipulated is by using magnetic field pulses. Ultrashort magnetic field pulses can be generated optically using circular polarized laser pulses with an energy smaller than the band gap. An ultrashort circularly polarized laser pulse can act as an ultrashort magnetic field pulse by the inverse Faraday effect[14]. The inverse-Faraday effect (IFE) is a Raman-like coherent optical scattering process by which a valance band electron with spin-up can end up, via virtual level in the bandgap with strong spin-orbit coupling, in a spin-down state in the valence band. In ferrimagnetic garnets films magnetic field pulses up to 0.6 T can be created[15]. The strength of the IFE is proportional to the magnitude of the magneto-optical Faraday effect at the used wavelength, which is large for EuO, and proportional to the intensity of the light. When 0.6 eV pulses are used, the absorption will be small and high intensities can be used without burning the sample. Since Eu-rich EuO has a negative CMR effect of six orders of magnitude[6], these magnetic field pulses are expected to have a dramatic effect on the conductivity. Again, possible changes in the conductivity can be probed by time-resolved THz-spectroscopy. Changes in the conductivity which result from an increased conduction band population can also affect the magnetic properties. The magnetization can be measured by time-resolved magneto-optical Kerr effect (MOKE) measurements.

Apart from addressing the nature of the switching phenomena, a study will be made towards the feasibility of using this material in spintronics applications by doing optically induced spin-precession and spin-grating experiments in the paramagnetic phase. Using circular polarized light it might be possible to induce a non-equilibrium spin population in the conduction band as was already demonstrated in GaAs. In GaAs the transition probability for a transition to a spin-up state in the conduction band is three times larger as the transition to a spin-down state, creating a spin imbalance. Now by applying a magnetic field perpendicular to the induced magnetization will result in precession. The magnetization can be probed by a time-resolved MOKE measurements. From the decrease of the Kerr signal in time, the spin-dephasing time can be obtained with can tell us something about the spin-dephasing mechanisms.

From spin-grating experiments the spin-lifetime and spin mobility can be measured which can be different from the electron lifetime and mobility as was shown in for GaAs quantum wells[16]. When two beams with parallel polarizations intersect on the sample, the light amplitude on the sample is not uniform. This creates a charge grating since on some places more electrons are excited to the conduction band than on other places. The disappearance of this grating in time can be monitored by measuring the amplitude of the diffracted probe pulse from this grating. When this is done for different grating periods, the electron mobility can be obtained.

When two beams with crossed-linear polarizations intersect, the light amplitude on the sample is uniform but the electric field polarization is spatially modulated across the excitation region. The polarization is alternating left and right circularly polarized across the excitation region with in between regions that are linear polarized. In this way we will try to make a spin-grating in EuO. The disappearance of this grating in time can be monitored by measuring the rotation of polarization of the diffracted

probe from the spin-grating. When this is done for several grating periods, the spin-diffusion and spin-relaxation time can be obtained. Also the spin-transport through the magnetization grating can be studied using combined optics and spin-polarized transport techniques to verify spin-filtering phenomena.
Before we can conduct all these experiments first good quality samples have to be made. The main problem in this is the formation of higher oxides (like Eu_2O_3) during the growth and the europium-oxygen stoichiometry. Control of the stoichiometry is crucial: Eu-rich EuO has a MIT but stoichiometric EuO does not have a MIT. At the university of Cologne in the group of L.H. Tjeng, the growth of EuO and Gd-doped EuO films is well under control. However never an EuO MOSFET is grown and a lot of efforts needs to be done to find the good parameters, like oxide material, oxide layer thickness, gate electrode material, contacts end so on. For the proposed project this group has an on going cooperation with the group in Köln. The grown samples can be characterized by SQUID measurements, UV-VIS spectroscopy, IR-spectroscopy, Raman spectroscopy and Kerr hysteresis measurements.

D. Plan of work

The project will take 48 months in total. Below, a time schedule in months is given:

- Getting acquainted with the experimental equipment and the material. (3)
- Thin film growth at the University of Cologne. (3)
- General characterization: Raman spectroscopy, SQUID measurements, UV-VIS spectroscopy, IR spectroscopy. (6)
- Optically-induced magnetic and conductivity switching by photo-doping, (optical) magnetic field pulses and laser heating. (9)
- Spin-precession experiments. (6)
- Spin-grating experiments. (6)
- Field-effect doping induced magnetism. (6)
- Stoichiometric dependence of switching. (3)
- Photoconductivity measurements. (3)
- Writing thesis. (3)

10 Infrastructure

The Optical Condensed Matter Physics group has a variety of optical setups from which the most relevant equipment is listed below:

- Time resolved Magneto-Optical Kerr effect setup (20 fs pulses) with a superconducting magnet ($0< B<8$ T) and a Ti:Sapphire cavity dumped laser. Also transport properties (photoconductivity) can be studied on this setup. (Repetition rate 100 kHz- 80 Mhz, photon energy 1.4-1.9 eV; 2.8-2.8 eV)

- 150 fs amplified system coupled to an optical parametric amplifier (TOPAS). On this setup the cryostat can be placed between a magnet ($0<B<100$ mT). (Repetition rate 1 kHz, photon energy 100 meV- 3.5 eV)

- THz-setup for time-resolved THz measurements.

- $Nd:YVO_4$ 532 nm CW laser coupled to a micro- Raman spectrometer (T64000- Jobin Yvon) with a CCD detector. This is a Raman set-up (10-9000 cm^{-1}).

- Time-resolved photoluminescence setup.

- UV-VIS-NIR absorption spectrometer (200-3000 nm) for initial characterization.

Additionally, all facilities and equipment of the Material Science Centre ^{plus} are available. EuO, Eu-rich and Gd-doped EuO thin films, can be grown on a MBE setup at the University of Cologne in the group of prof. dr. L.H. Tjeng at the Institute of physics II.

11 Application perspective in industry, other discipline or society

In the proposed research we tend to extend the knowledge about EuO, as an interesting spintronic material. We will try to control the magnetic and transport properties optically, electrically or by an applied magnetic field. Good control of physical properties is crucial when one want to make spin-electronic devices where EuO can be used as data storage medium or as spin-filter. Also spin-relaxation and spin-diffusion will be studied which is important for the characterization.
Apart from that, we are also interested in the speed at which we can switch the magnetic and transport properties. We want to do this in a non-thermal way, which is potentially much faster than the thermal switching. This is because of technological importance since it can lead to faster computers but is also interesting from a fundamental point of view. With different optical techniques we want to characterize the (ultrafast) optically induced effects.

12 References

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