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# Field and temperature dependence of the magnetization in ferromagnetic EuO thin films

**S Mühlbauer[1,2], P Böni[2], R Georgii[1], A Schmehl[3], D G Schlom[3] and J Mannhart[4]**

[1] Forschungsneutronenquelle Heinz Maier-Leibnitz, D-85747 Garching, Germany
[2] Physik Department E21, Technische Universität München, D-85748 Garching, Germany
[3] Department of Materials Science and Engineering, Pennsylvania State University, PA, USA
[4] Experimentalphysik VI, Elektronische Korrelationen und Magnetismus, Institut für Physik, Universität Augsburg, Germany

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**Abstract**

Ferromagnetic EuO exhibits a metal–insulator transition showing a very large colossal magneto-resistance. Recently, it became possible to grow epitaxial films of $EuO_{1-x}$ on Si with spin polarization above 90%. The direct integration of EuO with Si will allow the fabrication of model systems for studying devices in the field of spintronics. In order to determine non-destructively the magnetic properties of thin films of EuO on Si, we have measured the critical angle of reflection by using polarized neutrons. The results confirm that the magnetic moment in the films is consistent with the bulk value. In addition, we show that the change of magnetization is not caused by repopulation of domains but by domain rotation.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

The ferromagnetic semiconductor EuO [1] is the only known binary oxide that can be grown in a thermodynamically stable form in contact with silicon [2]. The conductivity can be matched to the conductivity of silicon by introducing oxygen vacancies [3, 4], which is a key necessity for coherent spin injection in spintronic devices [5]. Therefore, heterostructures composed of Si and EuO may be used as model systems for studying applications of devices in the field of spintronics, where besides the charge, the spin degree of freedom is also used to control the flow of conduction electrons.

Bulk EuO exhibits a ferromagnetic to paramagnetic transition at $T_C = 69$ K. Doping oxygen vacancies, $EuO_{1-x}$ shows a metal to insulator transition around $T_C$, which can be substantially shifted by applying an external magnetic field, yielding a colossal magneto-resistance effect over several orders of magnitude [6]. Due to the spin-splitting of the conduction band in lightly doped EuO a spin polarization at the contact between EuO and the transport medium (Si) of 100% is expected.

Until recently, research into systems containing EuO has been hampered because EuO is unstable when exposed to air, which transforms EuO to non-magnetic $Eu_2O_3$. Moreover, it was shown that epitaxial layers of EuO grown via a buffer layer of SrO exhibited a magnetic moment of only 4.7 $\mu_B$ [7], a value that is significantly lower than the bulk value of 7.0 $\mu_B$ [7]. Very recently, Schmehl *et al* [8] succeeded in growing epitaxial films of $EuO_{1-x}$ directly on Si with spin polarization over 90% as measured by means of Andreev reflection. Therefore it may become possible to develop within the well established Si technology a device for producing spin-polarized electrons for devices in the field of spintronics.

The $EuO_{1-x}$ films were deposited by reactive molecular beam epitaxy on thermally cleaned (001) Si and (110) $YAlO_3$ and have been protected *in situ* against degradation when exposed to air by a 130 Å thick capping layer of Si or other material (for details see supplementary information of [8]). X-ray diffraction scans showed that the structural properties were of exceptionally high quality, comparable to single-crystalline EuO [8].

While the magnetization of EuO grown on $YAlO_3$ has been measured by means of a superconducting quantum interference device (SQUID), yielding a saturation moment $\mu_{sat} = 6.7\ \mu_B$ per Eu atom and a coercive field $H_c = 60$ Oe

 

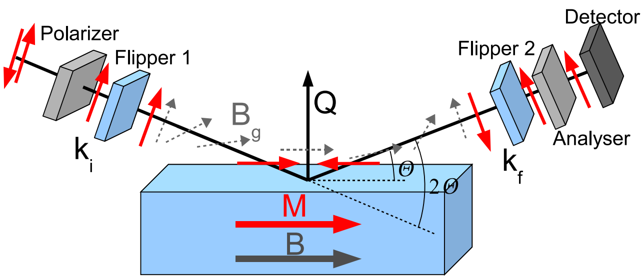


**Figure 1.** Schematics of the polarized neutron reflectometer MIRA. The incoming neutrons $\mathbf{k}_i$ are elastically reflected by the sample. $\mathbf{Q}$ designates the momentum transfer. The polarization of the neutron beam is selected by means of spin flippers before and after the sample. The polarization of the neutrons follows adiabatically the direction of the guide field $\mathbf{B}_g$. This setup allows the measurement of the four cross sections $I_{++}$, $I_{--}$, $I_{-+}$ and $I_{+-}$.

at 5 K [8], the magnetic moment of $EuO_{1-x}$ grown on Si has not been determined. The reason being that the large and rather elusive samples are supposed to be used for additional bulk measurements and should therefore not be reduced in size for measuring the magnetization with a SQUID. Moreover, bulk measurements would not provide information on the reorientation process of the domains, a property that is of importance for device applications.

The following notes provide a more detailed account of the determination of the magnetic moments in EuO films. The salient result $\mu_{\text{sat}} = 6.6\ \mu_B$ has already been published [8]. In order to determine the magnetic properties of the epitaxially grown $EuO_{1-x}$ films on Si we have measured the temperature and field dependence of the critical angle of reflection using polarized neutrons.

## 2. Experimental details

The reflectivity measurements were performed on the beamline for very cold neutrons (MIRA) at FRM II. The sample was mounted inside a closed cycle cryostat on a flat aluminium holder. A magnetic field was applied parallel to the incident neutron beam with wavelength $\lambda = 9.7 \pm 0.05$ Å and parallel to the sample surface, as sketched in figure 1. As the magnetic field is also guiding the neutron polarization, $\mathbf{P}$ is therefore parallel or antiparallel to the sample and perpendicular to $\mathbf{Q}$.

The amplitude of the magnetic moments can be determined by two means: (i) either by conducting measurements up to reasonably large $q$ and fitting the data with a bilayer model that includes the EuO and the capping layer or (ii) by determining the angle of total reflection that is given by [9] $\Theta_c = \lambda\sqrt{(\rho/\pi)(b_{\text{nuc}} \pm b_{\text{mag}})}$, which can be converted to a momentum transfer $q_c$ characterizing the drop of the intensity on a $q$-scale. Here, $\rho$ designates the density of the nuclei, $b_{\text{nuc}}$ the nuclear scattering length, and $b_{\text{mag}}$ the magnetic scattering length given by $b_{\text{mag}} = (\frac{\gamma r_0}{2}) g f(\mathbf{Q})\mathbf{S}$. Due to the low intensity of the MIRA beamline we have chosen method (ii) which is also used for the characterization of supermirrors. The reflection profiles $I_{++}$, $I_{+-}$, $I_{-+}$, $I_{--}$ were measured by performing $\Theta$–$2\Theta$ scans with fixed initial wavevector $\mathbf{k}_i$ for

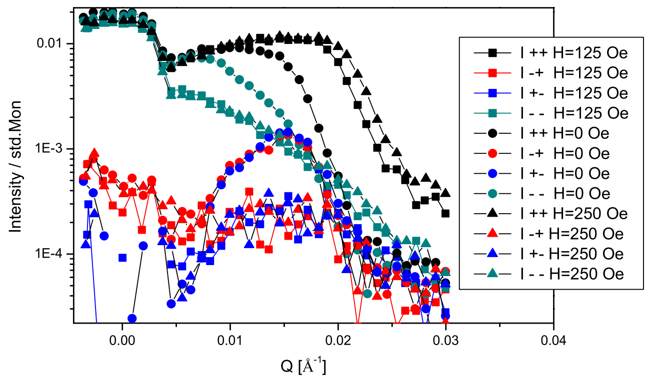


**Figure 2.** Reflectivity profile of EuO on Si(001), $T = 6$ K, $B = 0$, 125 and 250 Oe, full polarization analysis. The crosstalk between the individual channels due to finite beam polarization and flipper efficiencies has been corrected. The pronounced peak on the left-hand side identifies the unscattered direct beam, the dip between the plateau of total reflection and direct beam is due to footprint effects on the small sample. The critical angle of total reflection was determined at 50% reflectivity normalized on the total reflection plateau. Both spin-flip cross sections $I_{+-}$ and $I_{-+}$ vanish for $B = 125$ and 250 Oe while the data for $B = 0$ Oe show spin flip scattering.

temperatures 6 K $\leqslant T \leqslant$ 71 K in magnetic fields $B = 0$, 125 and 250 Oe.

## 3. Results and discussion

The obtained results can be summarized as follows: figure 2 shows a typical reflectivity profile of the EuO sample recorded at 6 K and fields $B = 0$, 125 and 250 Oe. The direct beam is visible around 0°. Due to footprint effects, the plateau of total reflection for $I_{++}$ is reached at roughly 0.011 Å$^{-1}$, while $I_{--}$ shows no total reflection. Due to the limited length of the sample, the total reflection plateau does not reach the full direct beam intensity. The critical angle of total reflection was determined at 50% reflectivity, normalized to the plateau of total reflection. The difference between

$$\Theta_{c1} = \lambda\sqrt{\frac{\rho(b_{\text{nuc}} + b_{\text{mag}})}{\pi}}$$

and

$$\Theta_{c2} = \lambda\sqrt{\frac{\rho(b_{\text{nuc}} - b_{\text{mag}})}{\pi}}$$

is clearly visible. The data have been corrected for a finite beam polarization of 92%.

The observed lack of spin-flip intensity for $B = 125$ and 50 Oe shows that the magnetization of the EuO is strictly parallel to the applied magnetic field, whereas the data recorded at $B = 0$ Oe clearly exhibit spin-flip scattering. This demonstrates that the change of magnetization is caused by domain rotation and not repopulation, ruling out $B \parallel M$.

Figures 3 and 4 show the temperature dependence of the reflectivity profile for the non-spin-flip cross sections $I_{++}$ and $I_{--}$ for $B = 125$ Oe. In figure 5, the temperature and field dependence of the critical angle of total reflection for

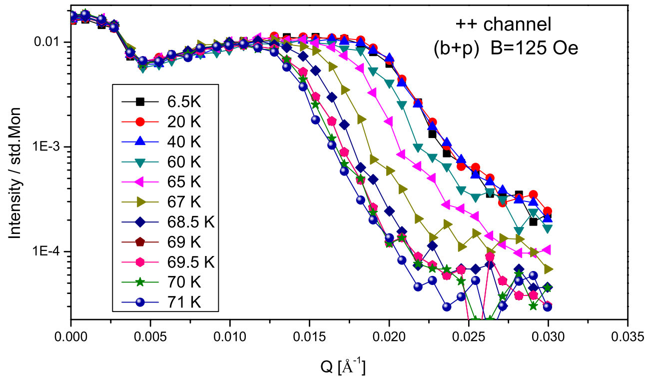


**Figure 3.** Reflectivity profile of EuO on Si(001) for temperatures from 6 K $\leqslant T \leqslant$ 71 K, $B$ = 125 Oe, $I_{++}$ cross section.

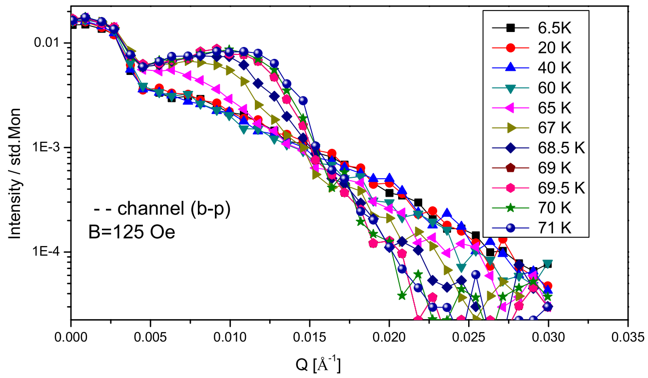


**Figure 4.** Reflectivity profile of EuO on Si(001) for temperatures from 6 K $\leqslant T \leqslant$ 71 K, for $B$ = 125 Oe, $I_{--}$ cross section.

$I_{++}$ and $I_{--}$ is shown. The merging of the critical angles for $T = T_c = 69$ K, where the ferromagnetism of the sample vanishes, is obvious. $M(T)$ exhibits qualitatively the expected decrease with increasing temperature [10]. A slight shift of the ferromagnetic to paramagnetic transition to higher temperatures for increasing magnetic field is visible, due to the field induced smearing of the ferromagnetic phase transition. The observed increasing magnetization with increasing field of the EuO layer is due to domain reorientation in the direction of the external applied magnetic field.

Two independent features of the measured reflectivity data can be used to verify the obtained values for $\Theta_c$.

The direct beam direction, defining $\Theta_c = 0$, has been used for performing an absolute calibration of $\Theta_c$. This yields a value $b_{mag}(Eu^{2+}) = 1.78 \pm 0.19 \times 10^{-12}$ cm for $T = 6.5$ K and $B = 250$ Oe, which is within 6.5% of the literature value ($b_{lit}(Eu^{2+}) = 1.904 \times 10^{-12}$ cm).

To cross check these results, the obtained data have also been normalized to the angle of total reflection of the nuclear contribution of $b_{nuc}(Eu^{2+})$ and $b_{nuc}(O)$, obtained above $T_c$, where no magnetic contribution is present, giving identical results for $b_{mag}(Eu^{2+})$. With $b_{mag} = (\frac{\gamma r_0}{2}) g f(\mathbf{Q})\mathbf{S}$ and $(\frac{\gamma r_0}{2}) = 0.2695 \times 10^{-12}$ cm a magnetic moment $\mu_{sat} = 6.6 \pm 0.7\ \mu_B$ is obtained per Eu atom, which is consistent with results from bulk single crystals.

Figure 3 shows the temperature dependence of the on-spin-flip cross section $I_{++}$. A flattening of the slope in the decay of reflectivity is visible for decreasing temperatures. This is due to a smearing of the EuO layer critical edge and the first order Bragg peak due to instrumental resolution. As the Bragg peak is less intense for decreasing magnetization, a flattening results for high temperatures. Due to the limited sample size of 2 × 1 cm$^2$, no off-specular data and no higher order Bragg peaks could be measured.

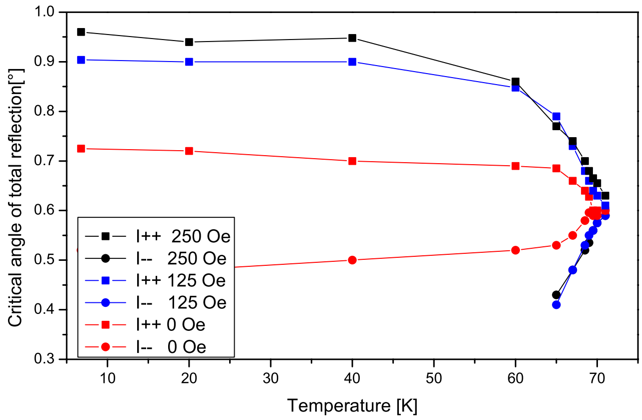


**Figure 5.** Temperature dependence of the critical angle of total reflection of EuO on Si(001) for different temperatures from 6 K $\leqslant T \leqslant$ 71 K and different applied magnetic fields $B$ = 0, 125 and 250 Oe. For $T = T_c$, the magnetic contribution vanishes. The clear field dependence indicates domain repopulation for increasing external fields in the direction of the applied external field.

## 4. Conclusion

By measuring the critical angle of total reflection with polarized neutron reflectometry, we demonstrated that the saturation moment of EuO grown epitaxially on Si(001) matches the moment of EuO grown on $YAlO_3$ as measured using a SQUID, without the need to cut the samples into small pieces. The observed magnetic moment of the $EuO_{1-x}$ layers agrees very well with values from bulk single crystals, proving the extraordinary high quality of the samples, whereas earlier measurements on EuO layers showed a reduced magnetic moment [7]. The samples clearly show a well defined ferromagnetic transition at $T_c = 69$ K. Thus, $EuO_{1-x}$, matched to the conductivity of silicon and grown epitaxially on silicon, may be broadly used for spintronic devices within the well established Si-based technology.

Furthermore, the sensitivity of polarized neutron reflectometry to the orientation of the magnetic moment in the sample proves that domain reorientation and not repopulation is responsible for the process of magnetization. Due to the limited sample size and the weak scattering intensity, no off-specular data could be collected and no higher-order Bragg peaks could be measured until now. Therefore, as yet no statements can be made with regard to surface and magnetic roughness or magnetization profiles.

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