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Polarized neutron reflectometry from the interface of the heterostructures $SiO_2(Co)/Si$ and $SiO_2(Co)/GaAs$

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ABSTRACT

Polarized neutron reflectometry is used to investigate $SiO_2(Co)$ granular films (70 at% of Co nanoparticles in SiO_2 matrix) deposited on Si and GaAs substrates. The aim of the study is to compare magnetization depth profiles in two systems: in $SiO_2(Co)/GaAs$ heterostructure which shows at room temperature giant injection magnetoresistance (IMR) with the system $SiO_2(Co)/Si$ which reveals almost no IMR effect. We found that at room temperature and at the same value of external magnetic field mean magnetization in the $SiO_2(Co)/GaAs$ sample is much higher than in the case of $SiO_2(Co)/Si$. We also demonstrate that magnetic scattering length density, and hence, magnetization profile strongly depends on the substrate. We show that $SiO_2(Co)/Si$ heterostructure is ferromagnetically ordered within the temperature range between 120 and 460 K what could explain a weak IMR.

1. Introduction

The interest in ferromagnet/semiconductor (FM/SC) interfaces is increasing last years due to the possibility of employing these structures as spin injectors of polarized electrons in SC [1–3]. Transfer of spin-polarized electrons (spin injection) from a magnetic contact into a nonmagnetic SC is intended for use in spintronic devices such as spin transistors, sensors, and magnetic memory cells [4,5]. The spin injection is noticed to depend on a potential barrier formed at the interface. Although significant progress has been made, the highly efficient spin injection into SC at room temperature remains a challenging task. As a solution we propose a FM/SC heterostructure where FM part is a granular film (GF) consisting of an insulator media with nano-size magnetic metal particles inside [6]. Electrons in the SC layer adjacent to the interface became polarized due to the RKKY-like exchange interaction with d-electrons in metal nanoparticles. Accordingly this interaction forms a potential barrier for spin-polarized electrons injected from the FM layer and enables their accumulation at the interface.

Recently, the giant injection magnetoresistance (IMR) in the heterostructure SiO_2(Co 60 at%)/GaAs was observed at room temperature [7,8]. The IMR effect is presumably caused by the suppression of the electron injection from the granular film into the GaAs substrate induced by the magnetic field and it is explained as surmounting the potential barrier by the injected electrons in the accumulation electron layer near the interface. The magnetic properties of the heterostructure SiO_2(Co 60 at%)/GaAs are supposed to be determined by the magnetic GF $SiO_2(Co)$ and by the spin-polarized accumulation electron layer in the SC.

The IMR effect depends strongly on the temperature. It is maximal at $T = 300$ K while it vanishes at low temperatures (smaller than 220 K) and at high temperatures (larger than 360 K). Fig. 1 shows the concentration dependence of the IMR effect observed at room temperature in the heterostructures on the GaAs and Si substrates. The magnetic field H was applied parallel to the film surface and was equal to 2 T. The magnetoresistance is defined as

$$IMR = \frac{R(H) - R(0)}{R(0)} = \frac{j(0) - j(H)}{j(H)}$$

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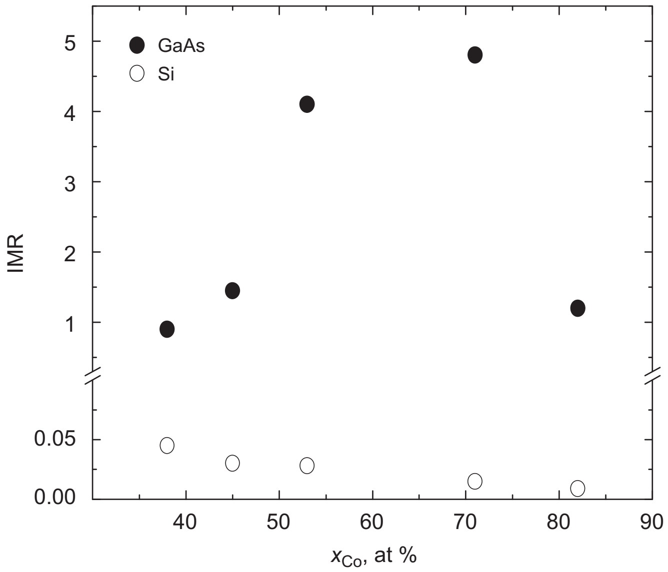

Fig. 1. IMR effect at $T = 300$ K and $H = 2$ T and $U = 60$ V in the heterostructures SiO_2 (Co x at%) on the GaAs and Si substrates.

where $R(0)$ and $R(H)$ are the resistances of the GF/substrate heterostructure, and $j(0)$, $j(H)$ are the injection current density in the absence and presence of the magnetic field H, respectively. As one can see the IMR effect depends strongly on the material of the substrate—it is very high for GaAs, while it is negligibly small in the case of Si. Therefore, the IMR effect has clearly of the interfacial origin.

In this study we used the polarized neutron reflectometry (PNR) for the investigation of the magnetic properties of a GF of SiO_2 containing 70 at% of Co nanoparticles on the Si and GaAs substrates. The study is focused on the evolution of magnetic states of the GF as function of the magnetic field at three temperatures of interest: $T = 120$, 300, and 460 K. These temperatures were chosen to provide different magnetic states of the film SiO_2(Co) which is, supposedly, ferromagnetic at $T = 120$ K, and 300 K and superparamagnetic at $T = 460$ K.

2. Samples

GFs SiO_2(Co) were prepared by the ion beam cosputtering of composite cobalt and quartz targets on the Si and GaAs substrates. In resulting products ferromagnetic Co nanoparticles are embedded into dielectric SiO_2 matrix. The average size of the Co particles estimated by means of small-angle X-ray scattering increases with the Co concentration from 2.7 nm for $x_{Co} = 39$ at% to 4.3 nm for $x_{Co} = 82$ at%. The granular structure of the SiO_2(Co) films deposited on the Si and GaAs substrates is equal. The thickness of the GFs is about 70 nm, the thickness of the Si and GaAs substrates is 0.4 mm.

3. Experimental technique

The experiments were carried out at reflectometer for polarized neutrons PNR (FRG-research reactor, GKSS Forschungszentrum Germany) that exploits a polarized neutron beam with initial polarization $P_0 = 0.95$ and neutron wavelength $\lambda = 6.35$ Å ($\Delta\lambda/\lambda = 0.05$) and reflectometer ADAM (ILL, France) with $P_0 = 0.95$ and $\lambda = 4.41$ Å ($\Delta\lambda/\lambda = 0.006$). The polarization of the incident beam followed the direction of the external magnetic field (from 0 to 320 mT at the sample position), which was applied perpendicular to the beam propagation vector and parallel to film surface. The reflected intensity was measured for two incident polarizations $I(+P_0)$ and $I(-P_0)$ along and opposite to the guiding magnetic field. The PNR method was used because it offers the possibility to resolve both magnetic and nuclear density depth profiles of thin films and multilayers deposited on a solid substrate [9].

4. Data evaluation and interpretation

The PNR data for every temperature point were taken after the demagnetization process i.e. after application of the opposite magnetic field up to 900 mT. No difference between I^+ and I^- components was observed for the sample SiO_2(Co 70 at%)/Si at $H = 0$ indicating zero net magnetization of the film. However, when external field was applied, the GF SiO_2(Co) magnetized what resulted in splitting of the I^+ and I^- reflectivity curves. Reflectivity profiles obtained for the sample SiO_2(Co 70 at%)/Si at $T = 120$ K and $H = 320$ mT are shown in Fig. 2. Both reflectivity components were fitted simultaneously with a 3-layer model scattering length density (SLD) profile every layer of which had four parameters—thickness, roughness, nuclear SLD N_n and magnetic SLD N_m. The latter has the opposite signs for $I(+P_0)$ and $I(-P_0)$ reflectivity components and value is directly proportional to the in-plane magnetization of an appropriate layer

$$M = \left(\frac{m_n \mu_n}{2\pi\hbar^2}\right)^{-1} \frac{N_m}{4\pi} = 3.44 \times 10^8 N_m.$$

Here m_n is the neutron mass, μ_n the neutron magnetic moment, N_m the magnetic SLD in Å^{-2}, and M the magnetization in Gs.

Fitting of two components together and relatively small experimental error bars allow for distinguishing some details of SLD distribution in the GF of total thickness 73.5 ± 0.5 nm for SiO_2(Co 70 at%)/Si and 56.5 ± 0.5 nm for SiO_2(Co 70 at%)/GaAs. In particular, one can see that magnetized part of the film is separated from both GF/substrate and GF/air interfaces by non-magnetized layers where SLD does not split.

Fig. 3 illustrates field dependences of magnetic SLD (normalized to magnetic SLD of bulk cobalt $N_{m(Co)} = 4.24 \times 10^{-6}$ Å^{-2}) of the middle part of the sample SiO_2(Co 70 at%)/Si at different temperatures. From Fig. 3 it

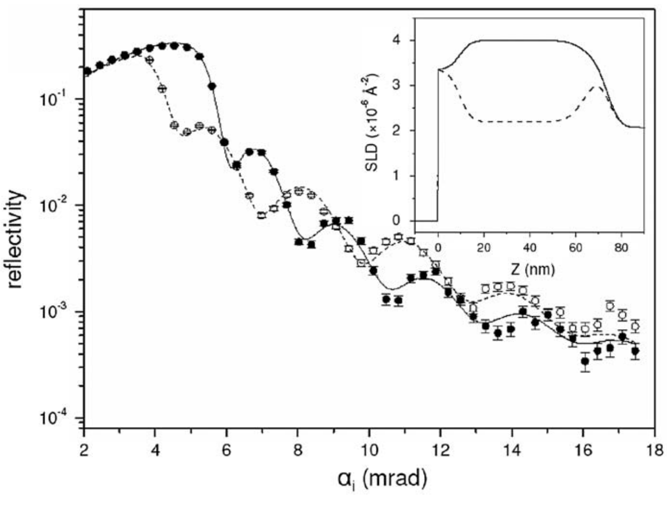

Fig. 2. Polarized neutron reflectivity data for the heterostructure SiO_2(Co 70 at%)/Si taken at $T = 120$ K and $H = 320$ mT. Experimental data are shown by filled (I^+) and open (I^-) circles, solid and dashed lines represent corresponding calculated reflectivities. The insert shows a 3-layer model SLD profile used for the fitting of experimental data—solid line for I^+ and dashed line for I^-.

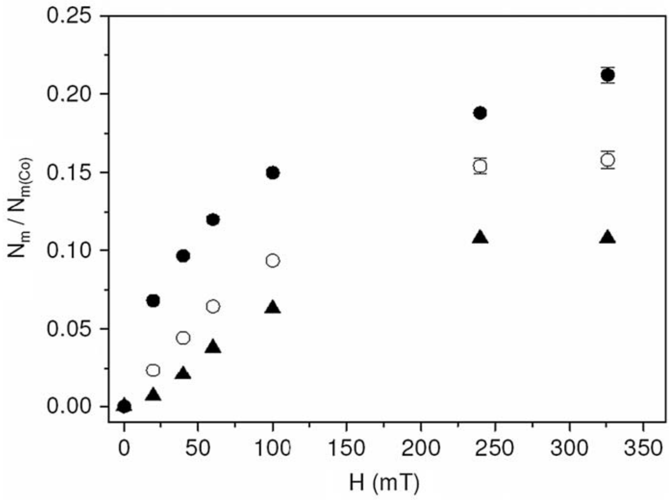

Fig. 3. Magnetic field dependence of fitted magnetic SLD value N_m for the ferromagnetic film SiO_2(Co 70 at%) as obtained at $T = 120$ K (filled circles), $T = 300$ K (open circles), and $T = 460$ K (triangles).

follows that the middle part of the GF is in the ferromagnetic state at all three temperatures. Its magnetization decreases with increasing temperature: at the maximum applied field $H = 320$ mT we found that $N_m/N_{m(\mathrm{Co})} = 0.212$ at $T = 120$ K, while decreasing down to 0.154 at $T = 300$ K and to 0.108 at $T = 460$ K.

Comparison of SLD profiles obtained for SiO_2(Co)/Si and SiO_2(Co)/GaAs at the same conditions $T = 300$ K, $H = 240$ mT is presented in Fig. 4. For the SiO_2(Co)/Si sample (Fig. 4a) one can not anymore identify strongly demagnetized intermediate layer between the GF and the substrate interface clearly seen in Fig. 2. Instead, one can recognize a "magnetically dead" top outmost layer and smooth reduction of magnetization towards a very sharp interface with the Si substrate. This is in contrast with the SLD profile in SiO_2(Co)/GaAs heterostructure. For this sample we also admit a very high magnetization (up to 0.59 $N_{m(\mathrm{Co})}$) of the GF and a slight signature of magnetization of the interfacial substrate layer. Although PNR results unambiguously demonstrate a distinct difference between magnetic and structural arrangements in SiO_2(Co)/GaAs and SiO_2(Co)/Si systems, it is too early to immediately link them to IMR effect. Further comprehensive studies, e.g. with PNR including full polarization analysis, X-ray surface scattering, etc., are required in order firstly to understand mechanism of dramatic effects of the interfaces on magnetization distribution over the films depth.

5. Conclusion

In summary, the presented results of PNR experiments show a dramatic influence of the substrate on magnetization depth profiles in SiO_2(Co 70 at%)/Si and SiO_2(Co 70 at%)/GaAs. It is, in particular, demonstrated that in the case of GaAs substrate at room temperature and $H = 240$ mT the magnetization value is a factor of 3 times higher than in the GF on the Si substrate. We found no influence of the external fields on the magnetic state of the GF in the heterostructure SiO_2(Co 70 at%)/Si substrate at all temperatures under study—it is always ferromagnetic. Absence of changes in the magnetic states correlates with negligibly weak IMR effect observed in the GFs on the given Si substrates.

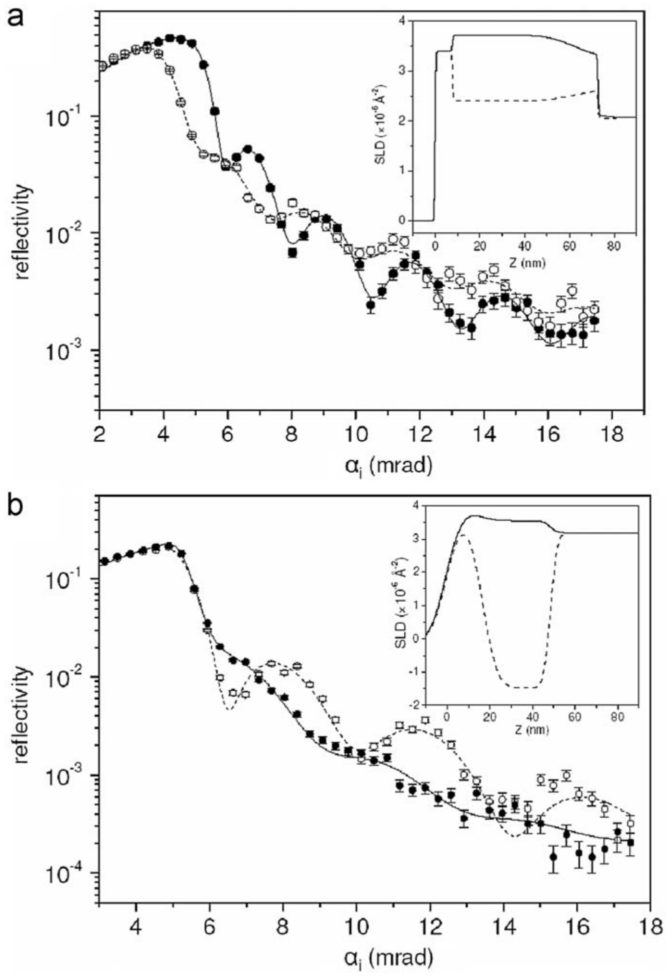

Fig. 4. Polarized neutron reflectivities obtained at $T = 300$ K and $H = 240$ mT for SiO_2(Co 70 at%)/Si (a) and SiO_2(Co 70 at%)/GaAs (b). All notations are identical to those of Fig. 2.

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