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Magnetic and Magnetotransport Properties of Magnetite/Co-Ferrite/Magnetite Trilayers

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The magnetic and magnetotransport properties of $Fe_3O_4/CoFe_2O_4/Fe_3O_4$ trilayers were investigated. The magnetization measurements indicate significant Co diffusion into the bottom magnetite layer. At lower temperatures the magnetoresistance measurements show two clearly separated maxima due to contributions from magnetically hard and soft layers. A tunneling effect was not observed. Comparison of full and minor resistance hysteresis loops did not reveal any significant exchange coupling between the layers, presumably due to diffuse interfaces.

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1. Introduction

Recently the spin-filter diode [1] has been developed as an alternative to the magnetic tunneling junction [2, 3]. In this device concept the tunneling barrier plays an active role in spin-polarizing the current passing between a ferromagnetic electrode and a non-ferromagnetic counter-electrode. Since the spin direction is conserved in direct quantum mechanical tunneling, the resistance of the spin-filter diode shows a strong dependence on the relative orientation of the magnetization vectors in the ferromagnetic electrode and the barrier [1, 4–6]. In this work a combination of conventional tunneling junction and spin-filter diode is investigated in the all--oxide $Fe_3O_4/CoFe_2O_4/Fe_3O_4$ system. The motivation in choosing ferrites lied in their predicted high spin--polarization [7, 8] and the high Curie temperatures of 858 K (Fe_3O_4) and 793 K ($CoFe_2O_4$) [9]. Moreover, there has been considerable work on ferrite heterostructures [10–12] and Co-ferrite has already been used as spin--filter in a $La_{0.7}Sr_{0.3}MnO_3/CoF_2O_4/Nb(0.5\%):SrTiO_3$ heterostructure [13].

2. Experimental

$Fe_3O_4/CoFe_2O_4/Fe_3O_4$ (FEO/CFO/FEO) samples were fabricated by pulsed laser deposition from stoichiometric Fe_3O_4 and $CoFe_2O_4$ targets onto MgO (001) substrates at 450°C in an oxygen partial pressure of 10^{-5} mbar. Just after the deposition of the bottom Fe_3O_4 layer, a mask was used to cover half of the substrate, and subsequently the $CoFe_2O_4$ spacer layer and the top Fe_3O_4 electrode were deposited. X-ray diffractometry measurements showed epitaxial growth of the magnetite layers, whereas the Co-ferrite spacer layer could not be resolved.

Here data on a sample with layer structure of 15 nm (top FEO)/8 nm (CFO)/60 nm (bottom FEO) are reported. Electrical contacts were made with silver paste and copper wires on the bottom and top Fe_3O_4 electrodes, respectively. The magnetoresistance was measured in a four-point out-of-plane configuration. Magnetization measurements were performed with a SQUID magnetometer. The diamagnetic contribution of the substrate was subtracted from the data.

3. Results and discussion

Figure 1 shows the magnetic characterization of the sample. The most striking feature is revealed by the lower panel showing magnetization hysteresis loops. From the structural data it is expected that about 10% of the sample contain the magnetically hard Co-ferrite phase. The magnetization data, however, show that this is not so, but that about 65% of the sample are magnetically hard. This can only be due to Co-diffusion from the Co-ferrite spacer layer preferably into the bottom electrode layer [14, 15]. Since this is a priori not unexpected, we have never observed this before in our work on $Fe_3O_4/Co_{0.16}Fe_{2.84}O_4$ bilayers [16]. Obviously the diffusion rate in the present case is considerably larger due to the high Co-concentration in the $CoFe_2O_4$ layer. In Fig. 1a the magnetization is shown as a function of tem-

perature. Below 150 K there is a significant difference between the magnetization curves measured in magnetic fields of 0.3 T and 1.0 T due to the temperature dependence of the coercive field of the hard layer. At about 100 K a small step-like anomaly can be seen in the magnetization curves; this is more clearly observed in the inset in the derivative $\mathrm{d}M/\mathrm{d}T$-curves. This might either be due to the Verwey transition or to the crossing of applied field and coercive field in the hard layer, $H_c(T) = H$. Since the anomaly — even if very faint — is still present in an applied field of 1 T with the sample already being in technical saturation, it is more likely to be caused by the Verwey transition of the top magnetite electrode.

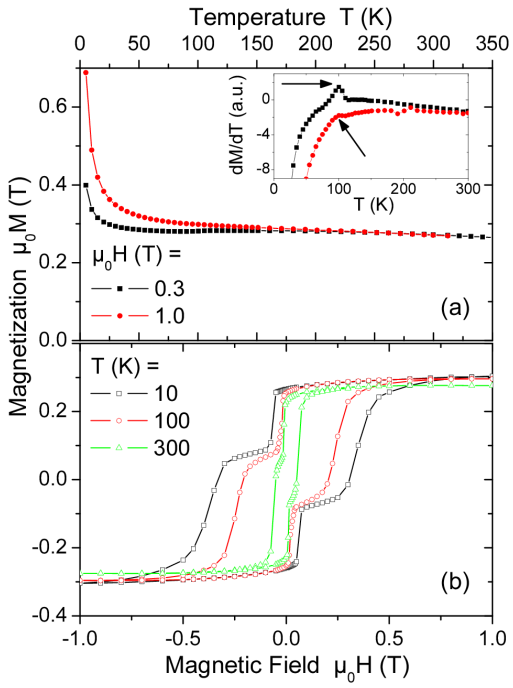

Fig. 1. (a) Magnetization of the sample as a function of temperature for applied magnetic fields of 0.3 and 1.0 T. The inset shows the temperature derivative of the magnetization. The arrows indicate weak anomalies in the derivative at 110 K. (b) Magnetization hysteresis loops measured at 10, 100 and 300 K.

Current–voltage characteristics measured at temperatures between 80 K and 300 K were linear. This precludes the observation of a tunneling current and tunneling magnetoresistance. In view of the strong Co/Fe-interdiffusion, this is not surprising. The resistivity of the bottom electrode was measured by a four point method. The resistivity shows clearly insulating behavior without any obvious resistivity jump that would reveal the Verwey transition.

The magnetoresistance (MR) $\mathrm{MR} = [R(H) - R(0)]/R(0)$ measured in an applied field parallel to the sample is shown in Fig. 2a for a temperature of 110 K. The MR clearly shows the typical butterfly contributions from both the magnetically soft and the hard phase. The maxima and shoulders of the MR occur at the coercive fields as indicated by the arrows. The MR is comparatively small and is certainly due both to anisotropic magnetoresistance and, since it shows a considerable high field slope, to grain-boundary magnetoresistance [16]. The general shape of the MR-curves does not change with temperature, the MR-value at 0.6 T decreases to −0.7% at 300 K.

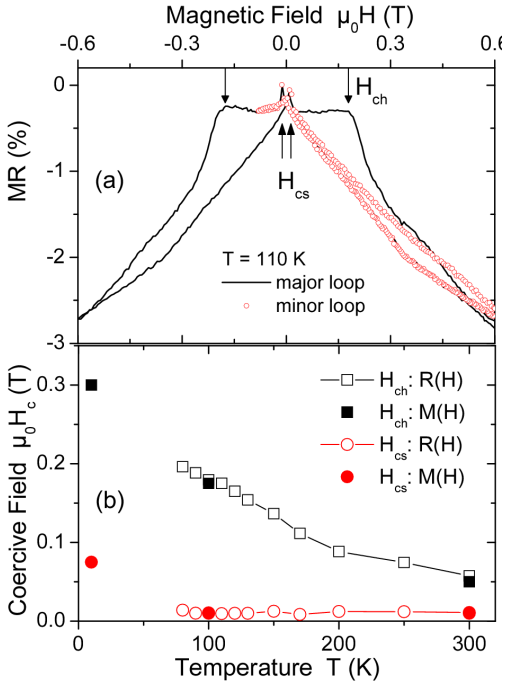

Fig. 2. (a) Full and minor resistivity hysteresis loops measured at 110 K. The magnetic field was applied parallel to the sample. The arrows mark the coercive fields of the magnetically soft (H_{cs}) and hard (H_{ch}) phases. (b) Coercive fields of the magnetically soft and hard phases as determined from resistivity and magnetization.

The magnetic coupling between the magnetically soft and hard layers was studied by magnetoresistance measurements. Besides the full magnetoresistance loop Fig. 2a also shows a minor loop. In the minor loop the magnetoresistance shoulder for positive magnetic fields is absent, since the hard layer was not reversed. However, no significant shift of the magnetoresistance maxima of the soft layer occurring at H_{cs} could be detected. This indicates that the magnetic coupling between the hard and the soft layers is weak. This is in contrast to the results on $Fe_3O_4/Co_{0.16}Fe_{2.84}O_4$ bilayers [17], which showed a strong coupling between the two layers. We believe that this is due to the absence of a sharp interface in the present samples due to the strong interdiffusion.

The coercive fields of the magnetically hard and soft phases were determined from the magnetization hysteresis loops and the maxima in the magnetoresistance hysteresis loops, respectively. The coercivity of the magnetite layer shows the generic temperature depen-

dence [18] being almost temperature independent between 100 K and 300 K and rising below 100 K due to the change of the magnetocrystalline constants at the Verwey transition, see Fig. 2b. The Co-doped magnetite layer shows a continuous rise in coercivity on cooling from 300 K down to 10 K in qualitative agreement with the results on $Co_{0.16}Fe_{2.84}O_4$ layers [17].

4. Conclusions

$Fe_3O_4/CoFe_2O_4/Fe_3O_4$ samples were fabricated by pulsed laser deposition and their magnetic and magnetotransport properties were studied. The magnetization measurements indicate strong Co/Fe-interdiffusion with the formation of a large fraction of a magnetically hard phase, i.e. Co-doped magnetite, in these samples. Tunneling conduction was absent. The magnetoresistance contained contributions from both phases due to anisotropic and grain-boundary magnetoresistance. The magnetic coupling between the magnetically hard and soft phases was weak, presumably due to diffuse interfaces. The coercive fields determined from magnetization and magnetoresistance hysteresis loops were in good agreement.

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