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Thickness dependence of ferromagnetic- and metal-insulator transition in thin EuO films

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We have studied the thickness dependence of the magnetic and transport properties of thin EuO films in the range of 10–60 Å. The ferromagnetic phase transition shows a systematic dependence of the critical temperature T_c with decreasing EuO film thickness. This behavior has been attributed to the interface layers which play a major role by reducing the number of average magnetic neighbors; we find the effect of interface intermixing becoming relevant in low thickness regime. In addition, we could identify a clear dependence of the onset of the metal-to-insulator transition on the ferromagnetic ordering of thin EuO films. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3063673]

Magnetic oxides have become an object of great fundamental interest in the field of spintronics because they can be effectively utilized as spin-filter materials.[1] Europium oxide belongs to the material class of rare earth magnetic semiconductors, and takes its magnetism from the half-filled 4f shell of the Eu^{2+} ion, giving rise to a spin-only magnetic moment of $S=7/2$. Especially challenging is to achieve sizable magnetic ordering and exchange splitting for film thicknesses of a few nanometers, which is the thickness regime of spin-filter tunnel barriers. Whereas recently various studies have been presented for bulk systems,[2,3] much less is known about the magnetic and transport properties of thin EuO films. The system raises several attractive fundamental questions, among which are dimensional effects in magnetism or spin transport.

In pure EuO, the ferromagnetic ordering is driven by the exchange coupling between the localized 4f moments of Eu^{2+} ions. Bulk EuO orders ferromagnetically at a Curie temperature of $T_c=69$ K. In a Heisenberg mean-field model, T_c is determined by the direct nearest-neighbor (NN) (J_1) and next-nearest-neighbor (NNN) (J_2) f-f interactions.[4] The transport properties of EuO are governed by the so-called $s(d)$-f exchange interaction between the localized Eu magnetic moments and the spins of the itinerant conduction electrons. The conduction band is split under the effect of exchange interaction, giving rise to a characteristic metal-to-insulator transition in the case of Eu-rich EuO. On the experimental side, the exchange splitting of thin EuO film was determined by Santos *et al.*[5] Theoretically, Schilling *et al.* studied the magnetic properties of ideal Heisenberg ferromagnets with $S=7/2$ in the monolayer regime.[6]

In this paper, we report a study of the thickness dependence of magnetic and transport properties of thin EuO films. Our experimental data clearly indicate the EuO film thickness as one of the primary factors scaling the ferromagnetic transition temperature and the onset of the metal-insulator transition. We show that the reduced magnetic ordering at interfaces due to structural and chemical intermixing becomes particularly relevant in the low thickness regime.

EuO film samples were prepared by thermal evaporation of pure Eu metal (99.9%) in an oxygen partial pressure of $p=(1-2)\times 10^{-7}$ Torr at room temperature. The films were grown on Si/SiO_2 substrates, with Al used as a seed and Y/Al as a top layer. Layer thickness and growth rate were monitored by a calibrated quartz monitor. *Ex situ* x-ray diffraction experiments of EuO films prepared under these conditions show a polycrystalline structure, with reflections originating from (200), (220), and (111) crystalline orientations.[7] The temperature dependence of the magnetization $M(T)$ was measured by superconducting quantum interference device magnetometry. T_c was determined as the temperature corresponding to the inflection point of the $M(T)$ dependence. For transport experiments, EuO was inserted as a tunnel barrier into cross-geometry tunnel junctions, yielding Al(4.2 nm)/EuO($d=1-4$ nm)/Y(2 nm)/Al(8 nm) stacks. The bottom Al electrodes were deposited at liquid N_2 temperature, whereas the EuO barrier and top electrodes were grown at room temperature, with an effective junction area of 200×200 μm^2. Electrical resistivity measurements were carried out by a four-terminal dc technique.

The temperature dependence of the ferromagnetic transition was studied as a function of EuO thickness varying from $d_{EuO}=10$ to 60 Å. Figure 1 displays the normalized low-field ($B=50$ Oe) magnetization $M(T)/M_S$ measured at 5 K, with M_S coming close to the bulk value of $6.9\mu_B$. The ferromagnetic transition temperature T_c thereby depends on the thickness of the EuO layer d_{EuO}. For structures with an EuO layer thicker than roughly 40 Å, the value of T_c is nearly identical (within the accuracy of T_c determination, which is ± 1 K) to the T_c of bulk EuO ($T_c\simeq 69$ K). With decreasing EuO film thickness $d_{EuO}<40$ Å, however, T_c is found to be shifted to lower temperatures with respect to bulk EuO. This result is in good agreement with Ref. 5.

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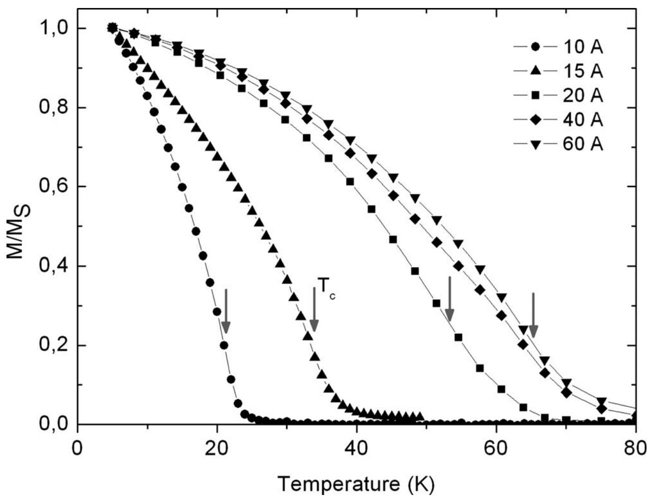

FIG. 1. Temperature dependence of the magnetization $M(T)/M_S$ at low field (B=50 Oe) for different EuO thicknesses d_{EuO}. Arrows indicate the ferromagnetic transition temperature T_c determined by the inflection point of the $M(T)$ curve.

We attribute the shift of critical temperature T_c to the reduced coordination number of magnetic neighbors located at the EuO interfaces. The magnetic Eu^{2+} ions in the outermost layers have less magnetic neighbors than their counterparts in fcc bulk EuO, which leads to a difference between the interface and center layer magnetization. Each Eu^{2+} ion is ferromagnetically coupled to its 12 NNs ($J_1/k_B = +0.606$ K) and its 6 NNNs ($J_2/k_B = +0.119$ K).[4] At interfaces, in contrast, the average number of NN and NNN is reduced depending on the crystalline orientation. Eu^{2+} ions in the outermost (100) interface layer have eight NNs and five NNNs, whereas in the (111) [(110)] orientation, the coordination number is reduced to six [seven] NNs and three [four] NNNs.

The effect of reduced dimensionality on the magnetic properties of a fcc(100) Heisenberg ferromagnet with $S=7/2$ has been investigated theoretically by Schiller and Nolting.[6] In Fig. 2, we compare our experimental results to their theoretical model of the layer-dependent transition temperature.[8] The experimental data roughly follows the overall tendency of the $T_c(d)$ dependence; however, a noticeable qualitative deviation between the experimental results and theoretical predictions can be observed. In particular, the Curie temperatures T_c of the experimental data are shifted to lower temperatures, especially for thinner EuO layers ($d \leq 20$ Å). This finding suggests that the short-range ferromagnetic NN and NNN coupling at interfaces of real substances is lowered compared to ideal systems.

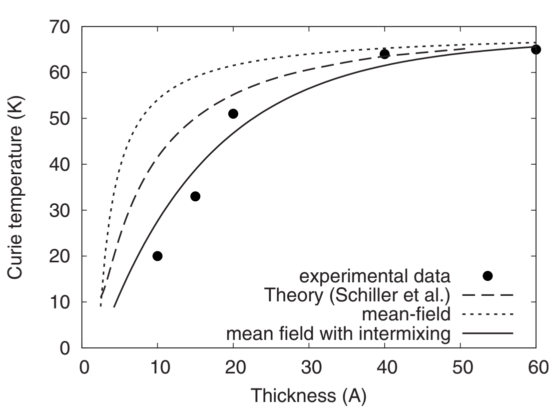

FIG. 2. Thickness dependence of the Curie temperature T_c. The dashed line corresponds to theoretical predictions for atomically flat interfaces by Schiller and Nolting (Ref. 6). Within a simple mean-field approach, an agreement with the experimental data is found by assuming a finite interface intermixing d_0 (solid line).

The theoretical description of the $T_c(d)$ dependence holds for atomically flat and free-floating interfaces. In the case of realistic EuO boundaries, however, imperfect interfaces can reduce the average number of magnetic neighbors even further. On the structural side, the presence of a finite interface roughness can lead to structural disorder, as the Eu^{2+} ions may be randomly distributed over the lattice sites of the cation sublattice. From a chemical viewpoint, magnetic Eu^{2+} ions can readily oxidize to nonmagnetic Eu^{3+} in the case of an oxygen surplus. It has been shown in Ref. 9 that the stable nonmagnetic compound Eu_2O_3 is mainly localized at the interfaces, and thus can account for the lowering of average short-range magnetic order. We believe that the effects of interface intermixing cannot be disregarded in our structures, as they very likely account for the lower Curie temperature T_c compared to theoretical predictions.

A simple analysis of the thickness dependence of T_c can be accomplished by a mean-field model, with $k_B T_c$ being proportional to the ferromagnetic ground state energy: $k_B T_c \sim \Sigma z_i J_i S^2$.[10] The transition temperature $T_c^{(n)}$ for n EuO monolayers scales with the average number of magnetic neighbors $\overline{z}_i$ according to $T_c^{(n)} = T_c^{\mathrm{bulk}}(\overline{z_{\mathrm{NN}}}J_1 + \overline{z_{\mathrm{NNN}}}J_2)/(12J_1 + 6J_2)$. Here, $\overline{z_{\mathrm{NN}}}$ and $\overline{z_{\mathrm{NNN}}}$ are the average numbers of nearest neighbors (coupled by J_1) and NNNs (coupled by J_2), which depend on the total number of EuO monolayers n. The differences between the surface and center layer coordination lead to the final expression for the layer-dependent magnetization $T_c^n = T_c^{\mathrm{bulk}}(1-c/n)$, where c is a numerical parameter. To account for the polycrystalline structure of the EuO films, we assumed an equal distribution of polycrystalline EuO grain orientations. We considered the effect of interface intermixing by introducing an attenuation factor $\propto \exp(-d/d_0)$, with d_0 being the characteristic scale of the EuO interface width.

In Fig. 2, we show the results of this semiquantitative approximation. Comparing the mean-field approach with and without interface correction, our analysis leads to a satisfactory agreement with the experimental data only, if a finite interface width $d_0 > 0$ is assumed. From the slope of the fitting curve, we can conclude that this effect becomes more relevant for small layer thicknesses. The fitting procedure gives an average interface width of about $d_0 = 12$ Å, which corresponds to about 4 ML. Assuming an initial roughness of the Al seed layer and taking into account the fractional formation of Eu_2O_3 according to Ref. 9, this result seems to be a reasonable approximation for the EuO structures. This finding is significant in particular for films below about $d = 20$ Å, whereas the influence of imperfect interface layers on the magnetic properties becomes less dominant for increasing film thickness. Our findings illustrate that the mean-field approach has the tendency to overestimate the ferro-

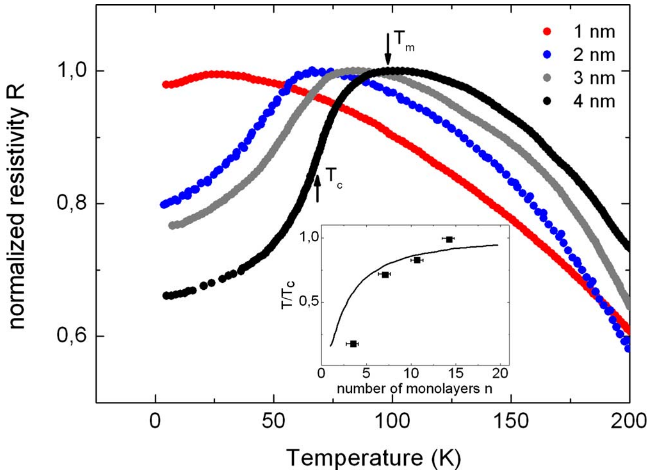

FIG. 3. (Color online) Temperature dependence $R(T)$ of the resistivity of Al/EuO/Y structures as a function of EuO barrier thickness. The onset of the metal-insulator transition shifts toward lower temperatures for decreasing EuO thickness. The inflection point at the low-temperature side of the $R(T)$ curve is identified with the Curie temperature T_c.

magnetic transition temperature in experimental systems[10] and that our simple model can describe the data more realistically by taking into account a reduced short-range magnetic order due to structural and chemical nonideal interfaces.

In the next step, we investigated the influence of reduced EuO layer thickness on the electrical transport properties. The electrical resistivity R of Al/EuO/Y structures was measured as a function of temperature T and EuO barrier thickness b, as shown in Fig. 3. For EuO barriers with $b=20-40$ Å, the characteristic resistivity dependence associated with a metal-insulator transition is observed, i.e., a sharp increase in the resistivity up to T_c, followed by a maximum at $T_m > T_c$ (see arrows in Fig. 3). As the thickness is reduced, $R(T)$ evolves toward a more insulatorlike dependence and the resistivity maximum reduces to a small feature superimposed on a monotonically increasing resistivity. Also, the temperature $T_m(R)$ at which the resistivity maximum is located decreases for smaller EuO thicknesses.

Our data show that the onset of the metal-insulator transition decreases with EuO layer thickness. In order to clarify whether this effect scales with the ferromagnetic order of EuO, we estimated the critical temperature T_c from the inflection point of the $R(T)$ curve.[11] This approach allows us to compare the variation in T_c obtained from electrical transport measurements to the theoretical results by Schiller and Nolting,[6] as shown in the inset of Fig. 3. We find that the tendency for $T_c(n)$ appears to qualitatively correlate with the theoretical predictions.

The driving mechanism for the metal-insulator transition is the exchange field created by the $4f$ electrons of the Eu^{2+} ions, which is reflected in the exchange splitting of the conduction electrons. The increasing influence of the EuO interfaces compared to the bulk contributions accounts for the reduced magnetic coupling between the magnetic ions in thin EuO films. In particular, this is expected to lead to a reduced exchange splitting of the conduction band, thus lowering the spin-filter efficiency for EuO films of few monolayer thicknesses. We thus may conclude that reduced short-range NN and NNN coupling of the localized magnetic ions can affect the transport properties of the conduction electrons in thin EuO films.

In conclusion, we consistently find the layer thickness of ferromagnetic EuO as one of the primary parameters scaling the magnetic and transport properties of thin film structures. Using a simple mean-field approach, we assigned the thickness dependence of T_c to a reduced magnetic coupling by taking into account layer-dependent magnetic moments and the effect of nonideal interfaces. Studying the transport in thin EuO films, we find the metal-insulator transition being lowered with decreasing EuO thickness and could relate this effect to the ferromagnetic state of thin EuO films.

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