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Modeling interlayer exchange coupling in EuS/PbS/EuS trilayers

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All-semiconducting EuS/PbS/EuS trilayers that show antiferromagnetic coupling were studied by superconducting quantum interference device magnetometry. We analyzed our measurements with a modified Stoner–Wohlfarth model from which the interlayer exchange energy and anisotropy were extracted based on the switching field from antiparallel to parallel alignment of the EuS layers and the zero-field susceptibility, respectively. Magnetic moment versus temperature curves were simulated by taking into account Brillouin type temperature dependence of the saturation magnetization of EuS. Despite their simplicity, the simulated curves show good qualitative agreement with the measurements when strong temperature dependence of interlayer coupling is assumed. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1676091]

In the future, magnetic semiconductor heterostructures may become crucial components of spintronic devices.[1,2] However, in contrast to the case of all-metallic structures, little is known about engineering the switching fields of magnetic semiconductor layers by exchange bias, or, alternatively, by exchange coupling to another magnetic semiconductor layer. Moreover, from a fundamental point of view, the origin of (interlayer) exchange coupling between magnetic semiconductors is still somewhat obscure and cannot be explained within the common Ruderman–Kittel–Kasuya–Yosida (RKKY) model for coupling in metallic systems,[3] since in semiconductors the carrier concentration is generally too low.

For the study of interlayer exchange coupling between magnetic semiconductors the EuS/PbS system is considered a suitable choice. EuS has a Curie temperature of 16.8 K, band gap of 1.6 eV and spin splitting of the conduction band 0.36 eV below T_C.[4–6] PbS is a nonmagnetic semiconductor, lattice matched to EuS with a band gap of 0.3 eV.[7] The magnetic properties and interlayer coupling of the EuS/PbS system have been the subject of several earlier studies, in which preferential antiferromagnetic alignment was observed at low fields and temperatures.[8–11]

An essential ingredient for a systematic study of magnetic properties is how to extract physical information like interlayer exchange energy or anisotropy from magnetostatic measurements. From a macroscopic point of view magnetic switching behavior is often described with the help of a Stoner–Wohlfarth model in which magnetic layers are treated as single domains.[12] This method has been applied successfully in the past to the study of magnetic properties in metallic structures.[13]

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In this article we discuss the use of a modified Stoner–Wohlfarth model that includes interlayer exchange to extract the exchange energy from magnetostatic data for all-semiconducting EuS/PbS/EuS trilayers. First the sample preparation and the measured magnetic properties are reviewed, followed by modeling of the magnetization curves based on a Stoner–Wohlfarth like model. Then, an extension of the model is proposed to describe the magnetic moment versus temperature measurements. We will argue that the observed magnetic behavior can only be explained by allowing the exchange coupling to vary strongly with the temperature.

EuS/PbS/EuS trilayers were grown epitaxially on freshly cleaved (100) KCl and PbS substrates by high vacuum evaporation of EuS using an electron gun and PbS with electrically heated tungsten boats at substrate temperatures of 250–300 °C. The trilayers consisted of two magnetic EuS layers of equal thickness, 30–200 Å, separated by a PbS spacer layer of 4–12 Å. The structures included 500–1000 Å thick PbS buffer layers to accommodate strain, as well as a 100–700 Å thick PbS cap layer to protect the trilayer from oxidation. We estimate that interdiffusion at the interface between EuS and PbS corresponds to intermixing of 1–2 monolayers (3–6 Å). We investigated around 40 samples on PbS substrates, and all showed in-plane cubic anisotropy.[9] The saturation magnetic moment is generally equal to the expected value for EuS of $7\mu_B$/atom within 5%–10%.

We will focus on one representative sample for all trilayers with PbS spacers between 4 and 12 Å which contains two 40 Å thick EuS layers separated by a 2.5 monolayer (7.5 Å) PbS spacer layer. Both a hysteresis curve and the magnetization versus temperature for different fields are shown in Fig. 1. At small applied fields a plateau of low total magnetic moment was observed, a feature we associate with mutual antiferromagnetic alignment of the two EuS layers. At a certain field, which we denote switching field H_s, the antipar-

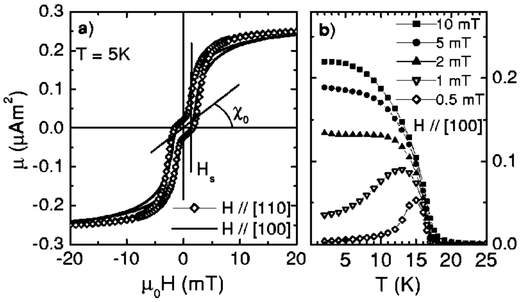

FIG. 1. (a) Easy axis hysteresis curve at 5 K for an EuS (40 Å)/PbS (7.5 Å)/EuS (40 Å) sample. H_s is the average switching field between parallel and antiparallel alignment and χ_0 is the zero-field susceptibility. The solid line represents a hard axis curve, which is not further discussed. (b) Magnetization vs temperature measurements along a hard axis.

allel configuration of the EuS layer magnetization is transformed into a parallel orientation. The field plays an important role in quantitative determination of the exchange energy, as we will demonstrate below. With respect to the temperature dependence of the magnetic moment, for small applied fields, the magnetization shows a sharp decrease in magnitude below a certain temperature as a consequence of a change from ferromagnetic (F) to antiferromagnetic (AF) alignment of the two magnetic layers [see Fig. 1(b)].

A Stoner–Wohlfarth-like model is introduced to quantitatively analyze the magnetization data. In the model, besides the magnetostatic energy and the fourfold (cubic) anisotropy, the interlayer exchange energy is also accounted for.[12] The total magnetic areal energy density E/A of two identical, single domain EuS layers with thickness t and saturation magnetization M is given by

$$E/A = -\mu_0 HMt\cos(\vartheta_1-\vartheta_H) - \mu_0 HMt\cos(\vartheta_2-\vartheta_H) + K_c t\sin^2\vartheta_1\cos^2\vartheta_1 + K_c t\sin^2\vartheta_2\cos^2\vartheta_2 - J\cos(\vartheta_1-\vartheta_2), \quad (1)$$

where the magnetic field applied is denoted by H, and ϑ_H, ϑ_1, and ϑ_2 are the angles between the field and the magnetization of each magnetic layer with a reference axis, respectively. The choice of direction for the reference axis (see the inset of Fig. 2) is, in principle, free, but it is taken along the field direction. K_c denotes the cubic anisotropy energy per volume unit, and J is the interlayer exchange energy per unit of surface area.

Magnetization curves are obtained by minimizing the areal energy density [see Eq. (1)] for each field applied with respect to the magnetization directions of the EuS layers. From the resulting angles the net magnetic moment of the structure along the reference axis is determined. We chose to minimize the global energy minimum, since Stoner–Wohlfarth models generally overestimate the coercivity, and that would also lead, in this case, to unrealistically large hysteresis.

Simulations of magnetization curves for the EuS (40 Å)/PbS (7.5 Å)/EuS (40 Å) sample are shown in Fig. 2 for several values of exchange energy and anisotropy. Figure 2(a) shows that, upon increasing the exchange energy, the width of the plateau of antiferromagnetic alignment becomes larger, i.e., the switching field becomes higher. The anisotropy determines the zero-field susceptibility, visible in the slope of the curve for small fields; see Fig. 2(b).

We selected as the best fit the simulation that has both switching field H_s as well as zero-field susceptibility χ_0 coinciding with those from the measured curve, indicated in Fig. 1(a). Because the plateau width is very sensitive to the exchange energy, the accuracy of the fit is mainly determined by the uncertainty in determining the switching field from the experimental curve. This is limited by the intrinsic nonideal behavior of the hysteresis loop of a single EuS layer, since the hysteresis curve of a single layer is as strained for high applied fields as the one of the trilayer. Similarly, accuracy in the determination of the anisotropy depends on the uncertainty in measured zero-field susceptibility, which will be overestimated if the two EuS layers do not have exactly the same magnetization. The fit thus provides a lower boundary for the anisotropy. The simulation with $J=-7\ \mu\text{J/m}^2$ and $K_c = 9\ \text{kJ/m}^3$ produces the best fit for the typical measurement shown in Fig. 1(a).

In order to simulate the temperature dependence of the magnetic moment, the temperature dependence of the saturation magnetization, anisotropy, and exchange also needs to be taken into account. The saturation magnetization of individual EuS layers generally follows mean-field behavior.[11] A detailed analysis of hysteresis curves measured at various temperatures suggests power-law dependence of both the anisotropy and the interlayer exchange coupling on the layer magnetization,[11,14] which we also applied in the present simulations.

Figure 3 shows the results of such simulations for the same sample with the magnetic field along a hard [100] axis. Figure 3(a) shows simulations for different applied fields. The simulations reproduce the qualitative behavior of the total magnetic moment: for high fields a monotonous increase with a decrease in temperature, and for relatively low fields an initial increase, followed by a decrease of the total magnetic moment, due to a change into antiferromagnetic alignment of the two magnetic layers. Variation of the critical exponent in the power-law dependence of J on layer magnetization M changes the general shape of the magnetic moment versus temperature curve and the temperature at which

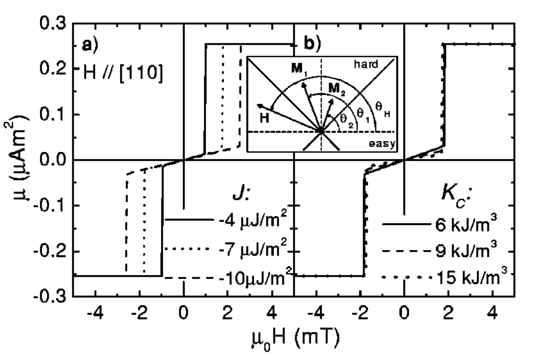

FIG. 2. Model calculations simulating the 5 K magnetization curve of the EuS (40 Å)/PbS (7.5 Å)/EuS (40 Å) sample for a magnetic field along the easy [110] crystal axis: the influence of (a) exchange energy J for $K_c = 9\ \text{kJ/m}^3$ and (b) anisotropy K_c for $J=-7\ \mu\text{J/m}^2$. The inset identifies the angles used in the model.

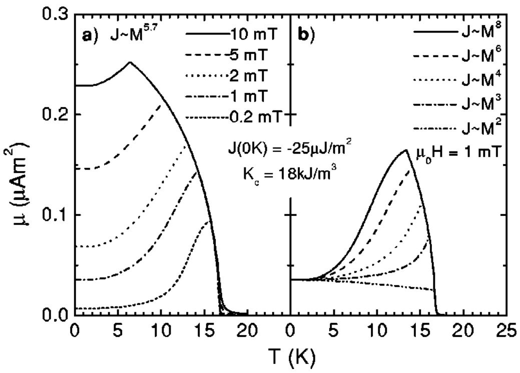

FIG. 3. Model calculations of the net magnetic moment as a function of the temperature for a EuS (40 Å)/PbS/EuS (40 Å) system with magnetic field along the hard [100] crystal axis: (a) the impact of magnetic field H and (b) the influence of the exponent in the expression $J\sim M^{\mathrm{exp}}$. Anisotropy energy K_c is taken to be $K_c^{\mathrm{eff}}=K_c(M/M_s)^4$ as reported previously (Ref. 11).

the maximum magnetic moment is reached; see Fig. 3(b). For $J\sim M^2$ a monotonous increase in magnetic moment is expected, from which we conclude that the experimental dependence of J on the magnetization/temperature is stronger; see Fig. 1(b). The mechanism behind the power-law behavior of $J(M)$ is not yet understood, but is currently being investigated.[14]

It should be emphasized that the present model can qualitatively describe the magnetic moment versus temperature dependence, but it cannot describe the exact quantitative behavior for all applied fields with the same parameters. The reason it cannot is probably the use of a Brillouin function to describe the temperature dependence of the magnetization. For small applied fields the magnetization of a single EuS layer is not saturated, since its magnitude depends on the field applied and on its orientation. Therefore the two EuS layers may have unequal magnetic moments if they are not parallel. An improvement would be to replace the Brillouin function with a more realistic function that could account for the dependence of the magnetization on the field and on their relative orientation.

Generally, simulations for different samples lead to exchange energies of the order of 1–50 μJ/m^2 at low temperatures, in fair agreement with neutron reflectivity data for spacer thicknesses above 7 Å.[8] However, for thinner spacers our values are lower than those from neutron reflectometry, a result we ascribe to ferromagnetic interlayer coupling via pinholes in the spacer layer. A detailed discussion of the physics of the interlayer coupling and its dependence on the temperature and nonmagnetic spacer thickness will be reported.[14]

Summarizing, antiferromagnetic alignment of the EuS layers was observed in EuS/PbS/EuS trilayers with PbS spacers of 4–12 Å. The interlayer coupling energy and the anisotropy were extracted for a PbS spacer layer thickness of 7.5 Å from simulations of the magnetization curves based on a modified Stoner–Wohlfarth model. Simulations of the magnetic moment versus the temperature indicate strong dependence of the interlayer coupling energy on the temperature or magnetization.

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