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Magnetic Properties of EuS/Co Multilayers on KCl and BaF₂ Substrates

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Magnetic and structural properties of EuS/Co multilayers were studied by magnetic optical Kerr effect and SQUID magnetometry techniques and by X-ray diffraction method. The multilayers containing monocrystalline, ferromagnetic EuS layer (thickness 35–55 Å) and metallic Co layer (thickness 40–250 Å), were grown on KCl (001) and BaF_2 (111) substrates using high vacuum deposition technique employing electron guns for Co and EuS. All investigated EuS/Co multilayers exhibit ferromagnetic properties at room temperature due to Co layer with the ferromagnetic transition in EuS layer clearly marked upon cooling below 16 K. In EuS/Co/EuS trilayers grown on KCl substrate the antiferromagnetic alignment of magnetization vectors of Co and EuS layers was experimentally observed as a characteristic low field plateau on magnetization hysteresis loops and a decrease in multilayer magnetization below 16 K. In Co/EuS bilayers the characteristic temperature dependent shift of magnetization loops was found due to exchange bias effect attributed to the CoO/Co interface formed by the oxidation of the top Co layer.

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1. Introduction

EuS/Co multilayers are hybrid structures consisting of a model Heisenberg ferromagnetic semiconductor EuS with the Curie temperature $T_c = 16.5$ K and

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a ferromagnetic transition metal Co with $T_c = 1388$ K. Recent investigations of these multilayers revealed variety of important spintronic effects, such as an antiferromagnetic interlayer coupling in EuS/Co multilayers [1] and a very efficient spin filtering by EuS electron barrier (energy gap near 1.6 eV) in Al/EuS/Co tunneling structures [2]. Additionally, in the studies in EuS nanocrystallites embedded in a Co matrix [3], an increase in EuS Curie temperature up to 160 K was observed. These effects are expected to originate from the presence of additional charge carriers in EuS in close proximity to Co interface, and also from exchange interactions between spin polarized electrons across EuS/Co interface.

In previous studies of EuS/Co layered structures, polycrystalline or amorphous Co and EuS layers were grown on glass or on Si substrate with Pt/Co metallic buffer layer [1, 4, 5]. In this work, we exploit the technological method developed for the epitaxial growth of EuS/PbS multilayers on KCl and BaF₂ substrates [6], in order to study the magnetic properties of EuS/Co multilayers with monocrystalline EuS layer. The quantitative analysis of the experimental results was performed using the Stoner–Wohlfart model for magnetization of multilayer structures.

2. Experimental

EuS/Co/EuS trilayers and Co/EuS bilayers were grown on freshly cleaved KCl (001) and BaF₂ (111) substrates using high vacuum deposition technique employing electron guns for Co and EuS. Epitaxial 200–600 Å thick PbS buffer layer was deposited on the substrates from heated tungsten boat. The layer thickness covered the range 35–55 Å for EuS and 40–250 Å for Co. The structural properties of the layers were examined by both standard θ –2 θ diffraction scan and low-angle X-ray reflectivity methods, revealing monocrystalline PbS buffer as well as bottom EuS layer. As no diffraction peaks were observed for Co layer, this layer as well as the EuS layer grown on top of Co are expected to be either polycrystalline or amorphous.

Magnetic properties of the layers were examined using magneto-optical Kerr effect (MOKE) and superconductor (SQUID) magnetometry techniques. The MOKE experiments were performed in longitudinal geometry using He–Ne laser as a source of linearly polarized light with the laser spot of about 0.5 mm in diameter. The angle of incidence of light on the sample was about 30 degrees. The standard lock-in technique with photo-elastic modulator operating at 50 kHz and a Si diode detector were used. Measurements of magnetic hysteresis loops were performed in temperature range T = 4-250 K and in external magnetic fields up to 2 kOe applied in the plane of the multilayer. The magnetic hysteresis loops were measured in different places over the 5 mm × 5 mm sample area revealing good macroscopic magnetic homogeneity of the examined layers. To obtain the absolute values of magnetization of EuS/Co multilayers SQUID magnetometer was also used.

The representative MOKE magnetization hysteresis loops of Co/EuS bilayers and EuS/Co/EuS trilayers grown on KCl substrate are presented in Figs. 1

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Fig. 1. (a) Magnetization hysteresis loops obtained by MOKE technique for Co/EuS bilayers grown on KCl substrate for samples with different Co layer thicknesses. (b) Temperature dependence of the *exchange bias field* $H_{\rm E}$ (defined in the figure).

and 2. In the case of Co/EuS bilayers (see Fig. 1a), the characteristic shift of the magnetic hysteresis loops is visible indicating the well known exchange bias effect [7]. The temperature dependence of the *exchange bias field* for samples with different Co layer thicknesses is shown in Fig. 1b. We assign this experimental finding to the exchange coupling between Co layer and an ultrathin (about 2 nm) top antiferromagnetic CoO layer known to spontaneously form on unprotected Co layers in air [8].



Fig. 2. Magnetization hysteresis loops (MOKE technique) of EuS(40 Å)/Co(130 Å)/EuS(40 Å) trilayers grown on KCl (001) and BaF_2 (111) substrates with 600 Å thick PbS buffer layer.

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In the case of EuS/Co/EuS trilayer samples, in which the Co layer is protected by top EuS layer, no exchange bias is observed but a characteristic low field plateau on magnetic hysteresis loops measured at 4 K is clearly visible (blacksquare curve in Fig. 2). Upon increasing temperature, this plateau vanishes for magnetic hysteresis loops measured at 16 K — i.e. at temperature close to the Curie temperature of bulk EuS. Such behavior was already observed in the case of previously studied magnetically coupled EuS/PbS [9, 10] and EuS/SrS [11] multilayers and can be interpreted as a result of antiferromagnetic alignment of magnetization vectors of Co and EuS layers.



Fig. 3. Temperature dependence of the magnetization (SQUID magnetometry) of EuS(40 Å)/Co(130 Å)/EuS(40 Å) trilayers grown on KCl (001). The external magnetic fields shown in the figure were applied in the plane of the layer along (010) crystal direction.

Figure 3 shows the results of the measurements of the temperature dependence of magnetization M(T) carried out for EuS/Co/EuS trilayer. In agreement with the M(H) analysis, a decrease in multilayer magnetization below 16 K for H < 120 Oe is observed indicative of an antiferromagnetic interaction between Co and EuS layers in this structure. In the case of EuS/Co/EuS trilayers grown on BaF₂ (111) substrates the shape of the magnetic hysteresis loops (see open-circle curve in Fig. 2) i.e. small coercive field and lack of any low-field plateau — again like observed for the EuS/PbS/EuS//BaF₂ structures [12], indicates either the presence of ferromagnetic interaction between EuS and Co layers or the lack of interlayer coupling in this case.

3. Discussion and conclusions

In order to verify the interpretation presented in Sect. 2, the modeling of the experimental M(H) dependence was performed in the Stoner–Wohlfarth-like

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model [13]. In this model, single-domain layers with a saturation magnetization M_i , thickness t_i and cubic magnetocrystalline anisotropy constants K_i are considered. Additionally, interlayer coupling J between the magnetic layers is taken into account. Thus, the total magnetic areal energy density E/A of the model exchange coupled bilayer system can be expressed as [9]:

$$E/A = -\mu_0 H M_1 t_1 \cos(\theta_1 - \theta_H) - \mu_0 H M_2 t_2 \cos(\theta_2 - \theta_H) + K_1 t_1 \sin^2 \theta_1 \cos^2 \theta_1 + K_2 t_2 \sin^2 \theta_2 \cos^2 \theta_2 - J \cos(\theta_1 - \theta_2),$$

where H is the applied magnetic field, θ_H , θ_1 , and θ_2 are the angles between the reference axis and the applied magnetic field and the magnetization vectors of each layer, respectively. The choice of the reference axis is free and usually corresponds to the direction of the easy magnetization axis. The first two terms describe the Zeeman energy of the layers in an external magnetic field. J represents the interlayer coupling energy per unit of area. The M(H) curve was simulated by numerical minimization of the total energy as a function of the magnetization direction of each layer. Since the Stoner–Wohlfarth model generally overestimates coercivities, we chose to simulate the unhysteretic M(H) curves, obtained in the calculations of global minimum of the total energy. By fitting the low-field behavior, the interlayer exchange coupling energy J (proportional to the magnetic field switching from antiferromagnetic to ferromagnetic configuration) and the crystalline anisotropy constants $K_{1,2}$ (influencing mainly the slope of the plateau) can be determined [9]. As a result of such modeling procedure the antiferromagnetic interlayer exchange coupling between EuS and Co layers was found for EuS/Co/EuS//KCl trilayer with simulated values of the interlayer exchange coupling energy J of the order of -0.1 mJ/m^2 , and cubic anisotropy constants $K_1 = 30 \text{ kJ/m}^3$ and $K_2 = 90 \text{ kJ/m}^3$ of EuS and Co layers, respectively. The obtained value of J is about one order of magnitude greater than that obtained previously for EuS/PbS/EuS structures with ultrathin PbS nonmagnetic spacer [9, 14]. This difference can be simply understood as in EuS/Co/EuS ferromagnetic layers are in a direct contact which corresponds to the limiting case of zero non-magnetic spacer thickness. Surprisingly, our modeling indicates the presence of magnetic anisotropy in polycrystalline (amorphous) Co layer. Due to the lack of direct X-ray diffraction (XRD) diffraction evidence, it can only be speculated that this behavior is induced by the crystal structure and surface morphology of the bottom (monocrystalline) KCl/PbS/EuS part of the multilayer.

In conclusion, two interlayer coupling phenomena were observed in our MOKE and SQUID magnetometry investigations of magnetic properties of the EuS/Co multilayers grown by high vacuum deposition on KCl and BaF₂ substrates. Firstly, strong *exchange bias* effect due to the exchange coupling between Co layer and an ultrathin top antiferromagnetic CoO layer is visible in the case of Co/EuS bilayers. Secondly, in the case of EuS/Co/EuS trilayers grown on KCl (001) characteristic low-field plateau in MOKE hysteresis loops were observed

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indicating antiferromagnetic coupling between EuS and Co layers. These observations have been confirmed experimentally by the analysis of non-monotonic M(T)dependence and numerical simulation of M(H) dependence in exchange coupled multilayers performed in the Stoner–Wohlfarth-like model. No experimental evidence for antiferromagnetic interlayer coupling was found for EuS/Co/EuS layers grown on BaF₂ (111) substrates.

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