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Kerr Effect Investigations of Magnetic Interlayer Interactions in EuS–PbS Multilayers

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Kerr magnetometry was employed to study the temperature dependence of magnetization and magnetic hysteresis loops in ferromagnetic EuS–PbS semiconductor multilayers in the temperature range $T = 3-35$ K at low magnetic fields $H \leq 150$ Oe. For EuS–PbS/KCl(100) structures with ultrathin non-magnetic PbS spacer of 1 nm, we observed a maximum on the temperature dependence of magnetization at low fields $H \leq 30$ Oe. For higher fields, we found for these structures a regular mean-field-like increase in magnetization with decreasing temperature. The same regular behavior was also found for EuS–PbS/KCl structures with thicker PbS spacer, as well as for all EuS–PbS/BaF_2(111) multilayers independently of spacer thickness. For qualitative interpretation of these findings, we consider two magnetic contributions to the total energy of EuS–PbS multilayers: the Zeeman energy and the antiferromagnetic interlayer exchange coupling between ferromagnetic EuS layers via diamagnetic PbS spacer.

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1. Introduction

EuS–PbS multilayers are layered ferromagnetic epitaxial structures composed entirely of semiconductor materials. Europium sulfide (EuS) is a model non-metallic Heisenberg ferromagnet — a member of the family of europium chalcogenides [1, 2]. Lead sulfide (PbS) is the well-known IV–VI compound semiconductor with a narrow energy gap. It is a diamagnet and serves as a "non-magnetic" spacer layer in the EuS–PbS multilayer structures. Both materials crystallize in cubic (rock salt) crystal structure with their lattice parameters a matching very well ($\Delta a/a = 0.5\%$). It permits the epitaxial growth of pseudomorphic EuS–PbS structures with an overall thickness exceeding 100 nm [3, 4]. Electronic structure of EuS–PbS multilayers can be described as multiple quantum well of PbS confined by the ferromagnetic barriers of EuS with a type I ordering of band edges at the EuS–PbS hetero-interfaces [3–6].

Magnetic properties of EuS–PbS multilayers grown epitaxially either on KCl(100) or BaF_2(111) substrates were examined in Ref. [7]. The measurements of magnetization (applying superconducting quantum interference device — SQUID), ac magnetic susceptibility, and ferromagnetic resonance (FMR) showed that the ferromagnetic transition in EuS–PbS multilayers is observed even for ultrathin EuS layers of only 2 monolayers (2 ML). The Curie temperature T_C was found to decrease with decreasing EuS thickness for layers thinner than about 10 ML = 3 nm [3]. The ferromagnetic transition temperature in EuS–PbS multilayers depends also on thermal stress caused by the differences between thermal expansion coefficients of the substrate and of the multilayer [7–9]. FMR measurements of magnetic anisotropy showed that the volume (shape) contribution to the anisotropy dominates the surface one for practically all thicknesses of EuS layer [7, 8]. Therefore, in EuS–PbS multilayers at low magnetic fields the magnetization vector lies in the plane of the layer.

EuS–PbS multilayers form a ferromagnetic–non-magnetic multilayer system with negligibly small carrier concentration both in ferromagnetic and in non-magnetic layer. Therefore, they constitute an excellent model system to study the mechanisms of interlayer exchange interactions via non-metallic spacers. These interactions proved to be of importance for both basic and applicational research on metallic magnetic multilayers [10]. They also play an important role in new semiconductor spintronic magnetotransport and magneto-optical structures [11]. In metallic ferromagnetic multilayers the interlayer exchange is brought about by the spin polarization of conducting carriers (an analog of the Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction) [12]. This mechanism is expected to be negligible in most of semiconductor structures due to the lack of the required very high concentration of carriers. A number of new mechanisms of interlayer exchange was proposed specifically for semiconductor systems indicating the possibility for *ferromagnetic* coupling via either electrons on shallow donor centers or by Bloembergen–Rowland type excitations over the energy gap [13–15]. However recently, in neutron diffraction and SQUID magnetization studies, the *antiferromagnetic* interlayer exchange interaction between EuS layers via thin PbS spacer layer was discovered in EuS–PbS superlattices grown on KCl(100) substrates [16–18]. As far as we know, this is the first all-semiconductor ferromagnetic system that shows antiferromagnetic interlayer coupling between the magnetic layers. For structures grown on $BaF_2(111)$ only ferromagnetic interlayer coupling was observed. A theoretical model explaining these experimental findings was proposed in Ref. [19]. In this model the interlayer coupling stems from the spin-structure dependent contribution to the total energy of EuS–PbS multilayer related to the exchange coupling between $^4f^7$ magnetic orbitals of Eu and s, p, and d orbitals of Pb, S, and Eu forming the valence and the conduction band of EuS and PbS. The aim of our work is to provide new experimental evidence for the interlayer exchange interactions in EuS–PbS structures using magneto-optical Kerr (MOKE) technique for the analysis of the temperature and the magnetic field dependence of magnetization $M(H,T)$. Since our work reports the first application of the Kerr magnetometry to EuS–PbS multilayers, we will also briefly address the important issues of spectral response and optical design of the structures.

2. Experimental

We studied magnetic properties of EuS–PbS multilayers and trilayers grown on two insulating transparent substrates, KCl(100) or $BaF_2(111)$. The typical multilayer consisted of 10 or 20 repetitions of a basic EuS–PbS bilayer with EuS thickness in the range $d_{\mathrm{EuS}} = 3$–8 nm and PbS spacer thickness varying between $d_{\mathrm{PbS}} = 0.8-10$ nm. The multilayers were grown on 50 nm thick PbS buffer layer using the method of vacuum evaporation of EuS and PbS on monocrystalline substrates. The growth took place at substrate temperature $T_{\mathrm{s}} \approx 250°\mathrm{C}$. The molecular fluxes were produced using electron gun for the EuS and a standard resistive heating for PbS. The thickness of the layers was monitored *in situ* by a calibrated quartz resonator, as well as checked post-growth by X-ray diffraction. The surface morphology was characterized by atomic force microscopy. We investigated both the multilayers terminated with the top EuS layer and with the top PbS cap layer. It is an important experimental factor because our measurements were carried out for the radiation energy $\hbar\omega = 2.1$ eV, i.e. in the spectral range of strong fundamental absorption in PbS layers (for PbS, the energy gap $E_{\mathrm{g}} = 0.3$ eV at low temperatures). Although we could detect the Kerr effect for PbS cap layer up to 10 nm, in practice, for MOKE studies one should select multilayers with very thin PbS cap layer $d_{\mathrm{cap}} \leq 2$ nm or without it. The characteristic temperature and magnetic field dependence of the Kerr rotation in EuS–PbS multilayers proves that the effect we observe is of magnetic origin and any non-magnetic contribution is negligible. The strong Kerr effect in EuS originates from the large magnetization dependent shift of the conduction band edge [1, 2].

The measurements of the longitudinal MOKE were carried out in the temperature range 3–35 K in liquid helium continuous-flow cryostat. The external magnetic field ($H \leq 150$ Oe) was applied in the plane of the multilayer. The angle of incidence of light on the layer was about 30 degrees. A halogen lamp was used for the light source. The polarization of electromagnetic radiation was modulated by a mechanical modulator at a frequency of about 60 Hz. For detection of reflected radiation we used photomultiplier applying standard lock-in technique. In order to establish the optimal experimental conditions we studied the spectral dependence of the Kerr effect in EuS–PbS in the wavelength range $\lambda = 500-825$ nm covering the fundamental electronic transitions in EuS. In agreement with the experimental data for bulk EuS crystals, we found two maxima of the Kerr rotation: for $\hbar\omega = 1.65$ eV (corresponding to the absorption edge of EuS), and for $\hbar\omega = 2.1$ eV (corresponding to the maximum of absorption in EuS) [1, 2]. In our measurements we used the wavelength $\lambda = 625$ nm which corresponds to the second maximum.

3. Discussion and conclusions

Two representative Kerr hysteresis loops of EuS–PbS multilayers are shown in Fig. 1. The EuS–PbS structure which is grown on $BaF_2(111)$ shows a much softer loop than that grown on KCl(001) substrates. The former shows a magnetic remanence of about 95%, whereas the remanence of the latter is only about 20%. The strong reduction of the magnetic remanence in EuS–PbS/KCl multilayers may result from the presence of the antiferromagnetic coupling between EuS layers. Since the detailed understanding of the shape of the magnetic hysteresis loops of single (non-coupled) ultrathin EuS layers is still lacking, no definite conclusion can be obtained based on this evidence only.

An important information concerning the magnetic properties of EuS–PbS multilayers can be drawn from the studies of the temperature dependence of the Kerr rotation. Figure 2 shows the Kerr rotation at different magnetic fields in $[EuS(3\ nm)/PbS(1.2\ nm)]_{10}$ and $[EuS(3\ nm)/PbS(2.3\ nm)]_{10}$, both of which were grown on KCl(001) substrates. While the sample with the thicker PbS spacer shows

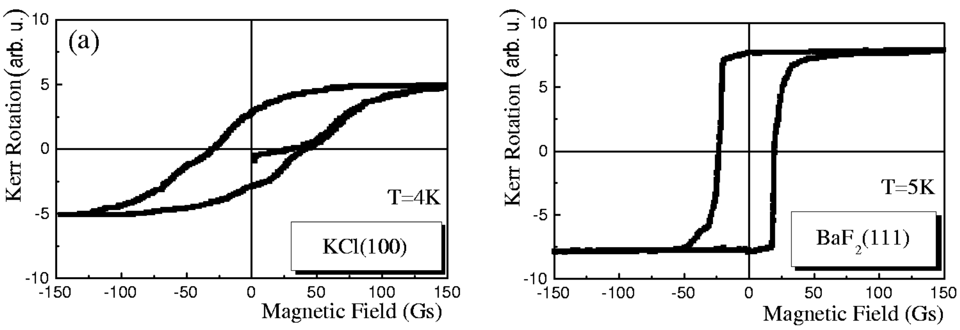

Fig. 1. The magnetic hysteresis loops in (a) [EuS(7 nm)–PbS(1 nm)] × 20 multilayer on KCl(100) substrate and (b) [EuS(7 nm)–PbS(1 nm)] × 20 multilayer on $BaF_2(111)$ substrate.

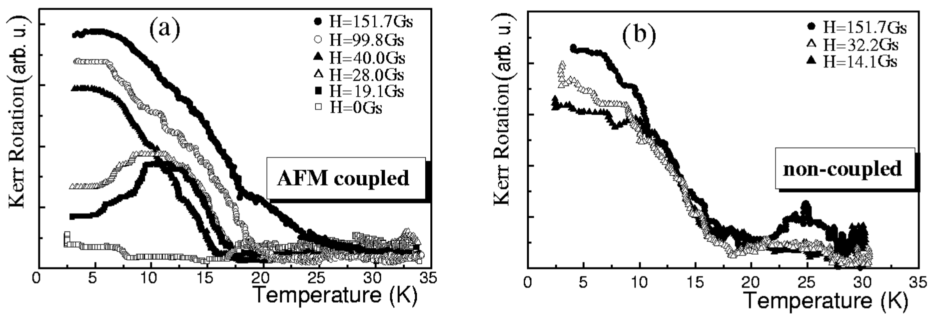

Fig. 2. The temperature dependence of the Kerr rotation in (a) [EuS(3 nm)–PbS(1.2 nm)] × 10/KCl(100) (thin PbS spacer), and (b) [EuS(4 nm)–PbS(2.3 nm)] × 10/KCl(100) (thicker PbS spacer). The measurements were performed applying in the plane of the layer, magnetic fields indicated in the figure.

the standard mean-field-like increase in the magnetization with the decreasing temperature, the sample with the thinner spacer shows an anomalous non-monotonic dependence of $M(T)$ in low external magnetic fields ($H \leq 30$ Oe). The "normal" $M(T)$ behavior is observed only in the presence of sufficiently high external magnetic fields ($H \geq 40$ Oe). These experimental findings can be explained in a simple model which considers the temperature dependent competition of the Zeeman energy of magnetic layers in external magnetic field (which favors ferromagnetic alignment of magnetization vectors of all EuS layers), and the exchange energy of antiferromagnetic interlayer coupling (which favors antiferromagnetic alignment of magnetization in coupled layers). Then, the decrease in the net magnetic moment with the decrease in temperature is a reflection that the different magnetic layers are becoming more strongly antiferromagnetically coupled. In fact, a simulation of the $M(T)$ curve with the inclusion of the antiferromagnetic interlayer coupling and using a mean-field Weiss theory for the description of magnetization of EuS layers can qualitatively reproduce the observed anomaly [17]. It is interesting to point out that no anomaly in the temperature dependence of the magnetization is observed in all the structures grown on $BaF_2(111)$ substrates, even for samples with thin PbS spacer layers ($d_{\mathrm{PbS}} \approx 1$ nm). This result is consistent with our suggestion concluded from the qualitative analysis of the Kerr hysteresis loops and favors the explanation that the high remanence in the structures grown on BaF_2 either indicates no interlayer coupling or the magnetic layers are ferromagnetically coupled. Another effect which must be taken into account in the analysis of the anomalous $M(T)$ behavior in the EuS layers is the temperature dependence of crystalline magnetic anisotropy. However, the contribution of this effect is of minor importance here because similar $M(T)$ behavior has also been observed in EuS–PbS multilayers on KCl(001) in both [100] and [110] in-plane crystallographic directions using SQUID magnetization measurements [17].

In conclusion, our study of the Kerr rotation of the EuS–PbS multilayers shows the presence of antiferromagnetic interlayer coupling in films which were grown on KCl and not those on BaF_2. This is in agreement with the recent study of the interlayer coupling in EuS–PbS by Kępa et al. using neutron scattering and neutron reflectivity techniques, as well as SQUID measurements of magnetic hysteresis loops [16]. Although the MOKE hysteresis loops do not show step-like behavior, indicative of the presence of interlayer coupling, the temperature dependence of the magnetization of the thinner PbS spacer sample does show a strong decrease in the net magnetic moment with decreasing temperature in low magnetic fields. This anomaly can be explained as a result of the competition between the Zeeman energy of ferromagnetic layers in external magnetic field and the temperature dependent contribution of the antiferromagnetic interlayer exchange energy. Our experiments show that the measurements of $M(T)$ curves can be a sensitive way of studying the antiferromagnetic interlayer coupling in semiconductor magnetic multilayers.

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