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Direct evidence for significant spin-polarization of EuS in Co/EuS multilayers at room temperature

S. D. Pappas[1], P. Poulopoulos[1,2,3], B. Lewitz[2], A. Straub[2], A. Goschew[2], V. Kapaklis[4], F. Wilhelm[5], A. Rogalev[5] & P. Fumagalli[2]

[1]Laboratory of High-Tech Materials, School of Engineering, University of Patras, 26504 Patras, Greece, [2]Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin-Dahlem, Germany, [3]Materials Science Department, University of Patras, 26504 Patras, Greece, [4]Department of Physics and Astronomy, Uppsala University, Box 516, SE-751 20 Uppsala, Sweden, [5]European Synchrotron Radiation Facility (ESRF), B.P.220, 38043 Grenoble, France.

The new era of spintronics promises the development of nanodevices, where the electron spin will be used to store information and charge currents will be replaced by spin currents. For this, ferromagnetic semiconductors at room temperature are needed. We report on significant room-temperature spin polarization of EuS in Co/EuS multilayers recorded by x-ray magnetic circular dichroism (XMCD). The films were found to contain a mixture of divalent and trivalent europium, but only Eu^{++} is responsible for the ferromagnetic behavior of EuS. The magnetic XMCD signal of Eu at room temperature could unambiguously be assigned to magnetic ordering of EuS and was found to be only one order of magnitude smaller than that at 2.5 K. The room temperature magnetic moment of EuS is as large as the one of bulk ferromagnetic Ni. Our findings pave the path for fabrication of room–temperature spintronic devices using spin polarized EuS layers.

Magnetic semiconductors are a relatively new class of promising materials for spintronic devices, like for example the fabrication of spin filters, spin diodes, spin transistors, spin qubits and non volatile magnetic memories[1–4]. Therefore, the search for ferromagnetic semiconductors and stable half-metallic ferromagnets with Curie temperatures (T_C) higher than room temperature (R.T.) is essential for Solid State Physics and Materials Science[5]. However, to our knowledge, a clear ferromagnetic response of semiconductors at R.T. has never been observed. Europium sulfide (EuS) is a natural ferromagnetic semiconductor. As compared to layered systems ferromagnet/semiconductor, it presents the advantage that the spin-polarized electrons are created within the semiconductor itself. The main disadvantage of it is a low T_C of only ∼16.6 K. In 1998, Fumagalli *et al.* reported that EuS nanospheres with a diameter of about 10 nm become ferromagnetic at about 160 K when placed in a Co matrix[6]. Since then, a strong effort has been put on making EuS ferromagnetic at R.T. by formation of EuS/Co trilayers with EuS layers thicker than 3.5 nm, but no promising direct evidence was provided[7–9]. Furthermore, it has been reported that pressure of the order of 90 GPa may bring the T_C of EuS close to R.T.[10].

In this work, we demonstrate a strong spin polarization of EuS at R.T. due to proximity to Co in two Co/EuS multilayers with 4 nm and 2 nm thick EuS layers. The polarization is approximately inversely proportional to the EuS layer thickness. Both samples show a mixed Eu valence state which does not seem to influence the magnetic properties as only Eu^{++} contributes to the magnetization. Element-specific magnetization curves show that the Eu magnetic moment is antiparallel polarized to the one of Co at remanence and rotates towards Co with increasing magnetic field. However, an applied magnetic field of even 17 T at low temperatures is not sufficient to orient the Eu moment parallel to that of Co for the sample with the thinnest EuS layers. At R.T., EuS remains antiparallel to Co for fields between 0 and 0.65 T indicating ferromagnetic ordering within the EuS layer . The strong spin polarization observed at R.T. results in a considerable magnetic moment which is comparable, for example, to that of ferromagnetic Nickel. This observation renders EuS layers thinner than about 2 nm in 3d metal/EuS trilayers and multilayers useful for R.T. spintronic applications. Moreover, since EuS is a semiconductor with a direct band gap at about 1.65 eV, combination of magnetic properties at R.T. with band gap tuning due to quantum confinement, which we have recently demonstrated, may merge spintronics and optoelectronics, as for example in spin-controlled light emitting diodes or vertical-cavity surface-emitting lasers[11].
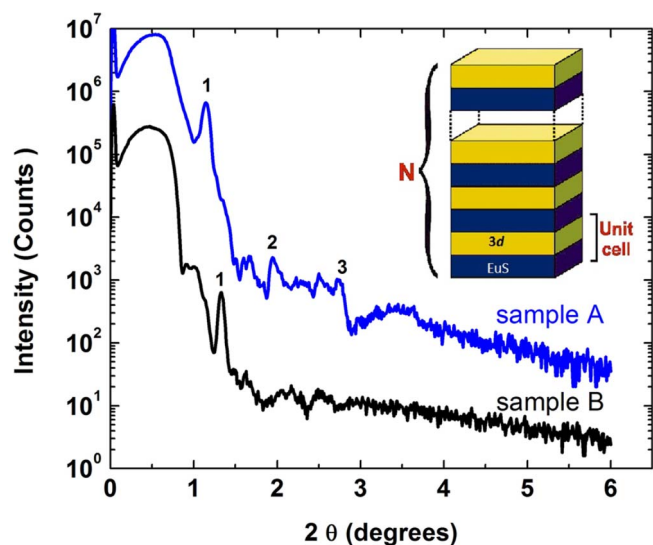
Figure 1 | X-ray reflectivity patterns for two Co/EuS multilayers grown on the native amorphous oxide of a Si(100) wafer. The pattern of sample A has been vertically shifted for clarity. In the inset, a typical multilayer structure between EuS and a 3*d* metal (here Co) is shown.

Results

Figure 1 plots the x-rays reflectivity (XRR) patterns of our multi-layers. In the first multilayer (sample A), three equispaced Bragg peaks are visible while in sample B, only one can be detected. This shows the formation of a multilayer structure. No Kiessig fringes appear due to relatively large total thickness and some roughness of the multilayer stack[12]. With the help of the XRR patterns and a precalibrated quartz balance, the layer thicknesses for the two samples A and B were found to be Co(7 nm)/EuS(4 nm) and Co(7 nm)/ EuS (2 nm), respectively. The number of bilayer repetitions was 9 and 15, respectively.

We present the main finding of our work in Fig. 2: Clear XMCD spectra at the L_2-edge of Eu are plotted for samples A and B at R.T. under an applied magnetic field 0.65 T. The magnitude of the spectra scales approximately inversely proportional to the thickness of the EuS layers. The external field is too small to account for this polarization. The significant spin polarization of Eu in EuS is attributed to
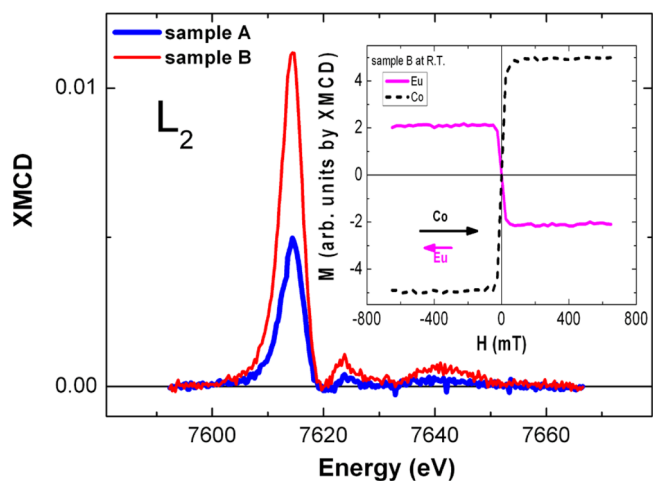
a strong exchange coupling due to the proximity to the Co layers. In the inset, we present element-specific magnetization curves at R.T. for sample B. We observe that the Co and Eu moments are always antiparallel. This is a clear indication of magnetic ordering within the individual EuS layers as the Co stray-field would always lead to a parallel alignment between Co and Eu. No significant change of the magnitude of the magnetization of each element occurs up to 0.65 T further corroborating magnetic ordering within the EuS layers.

In Fig. 3, the x-ray absorption (XAS) and the XMCD spectra at the $L_{3,2}$-edges of Eu are plotted for sample B under a field of 17 T at 2.5 K. This temperature is well below the T_C of bulk EuS. First of all, one has to notice that the white line of Eu at both edges shows two peaks. The first (second) peak originates from transitions between the 2*p* and 5*d* band of divalent (trivalent) Eu. In the same Figure, the XAS spectrum at the Eu L_2 edge for sample A has been introduced for comparison. It also shows two peaks but the first is more pronounced than the second.

The problem of valency of Eu in chalcogenides and other compounds of Eu is well known[13]. Trivalent Eu may originate from defects[14,15], pressure[16–18], or high magnetic fields[19]. For our samples, we did not observe any change of XAS by applying high magnetic fields, while no pressure was applied during measurements. However, growth of EuS at temperatures below 400°C is known to produce defects and, consequently, Eu^{+++} [14]. Therefore, we should attribute the presence of trivalent Eu mainly to defects. Residual strain at the interfaces with Co could also produce pressure at the interfaces and could be another source of Eu^{+++}. When the ratio of Eu^{++} and Eu^{+++} is close to 1/2 then Eu_3S_4 with cubic Th_3P_4 structure may form[13,20]. While Eu^{++} in EuS has a net spin magnetic moment of 7 μ_B/ion (J = 7/2), Eu^{+++} in Eu_2S_3 should have a zero magnetic moment because J = 0. Actually, trivalent europium was found to exhibit Van Vleck paramagnetism in europium nitrides[15]. Interestingly, according to Antonov et al.[21], the same holds for Eu_3S_4, i.e., only the Eu^{++} ions carry a magnetic moment which is equal to 7 μ_B/ion. This allows us to treat the magnetism of EuS in our samples as a problem of mean value and neglect the real crystallographic structure. We estimated the valency based on the relative weights of the integrated signal below the absorption lines normalized to the
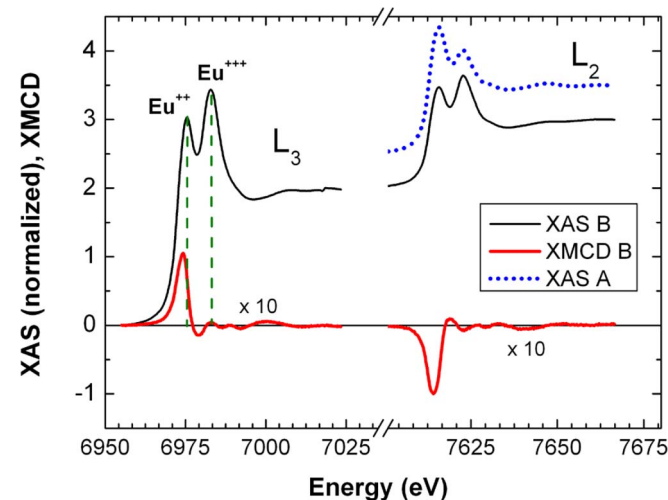

Figure 2 | Normalized XMCD spectra recorded at R.T. at the Eu L_2 edge for samples A and B. Significant polarization of Eu in EuS is revealed. Inset: Element-specific magnetization curves by XMCD at R.T. for sample B. Eu is always antiferromagnetically oriented to Co.

Figure 3 | Normalized x-ray absorption (thin solid line) and x-ray magnetic circular dichroism spectra (thick solid line) at the Eu $L_{3,2}$-edges for sample B at 2.5 K and 17 T. Two vertical dashed lines at the positions of Eu^{++} and Eu^{+++} are guides to the eye showing that the XMCD signal is concentrated at the divalent Eu. Finally, the dotted line represents the normalized XAS recorded at the Eu L_2-edge of sample A showing less trivalent Eu than sample B; the spectrum has been vertically shifted for clarity.

number of *d*-holes, after the subtraction of a double step-function, which accounts for the transitions to the continuum[18,22,23]. We found that sample A has about 63% Eu[++] and sample B 45% of the total amount of Eu. Therefore, the average magnetic moment of Eu in sample A should be 4.4 μ_B/ion and in sample B 3.15 μ_B/ion.

The intense XMCD signal is evidence of the presence of Eu magnetic moment. By knowing the direction of the magnetic field and the helicity of the beam, we conclude that the Eu magnetic moment projection on the 17 T magnetic field direction is positive at 2.5 K. Practically all the XMCD signal is concentrated at the $L_{3,2}$-edges of divalent Eu. Quantification of XMCD at the $L_{3,2}$-edges of Eu is impossible due to mixing of dipole (2p→5d) and quadrupole (2p→4f) transitions and lack of sum rules. XAS and XMCD recorded at the Co K-edge under the same experimental conditions revealed that (i) Co is in a hexagonal closed packed (*hcp*) structure and (ii) the magnetic moment of Co is always parallel to the magnetic field.

In Fig. 4, we present the XMCD spectra of both samples at the L_2 edge of Eu, which is a direct measure of the projection of spin magnetic moment of the ideal Heisenberg ferromagnet EuS on the external field. For sample B, one may observe that the strongest XMCD signal is not at 17 T but at remanence. This may be interpreted with the help of the element-specific hysteresis loops shown in the inset of Fig. 4. Co is always oriented along the external field. At large magnetic fields, Eu tends to orient along the field direction in order to minimize the Zeeman energy. However, the element-specific hysteresis loop is far from saturation, i.e., the Eu moment is still displaced from the field direction by a significant angle. A very strong direct antiferromagnetic coupling does not let Co and Eu to be fully parallel even under large external fields. The antiparallel (antiferromagnetic) alignment between the magnetic moments of transition metal ferromagnets and rare earths is documented experimentally and theoretically, see e.g. Ref. 24,25. Actually, the 5*d* states of the rare-earth owe their ferromagnetism to the polarization by the 4*f* states. The 5*d* states of the rare earth hybridize then with the 3*d* states of the transition metal giving rise to indirect exchange interaction with the 4*f* states of the rare earth element, see for example Ref. 25. The magnetization curve of Eu at 2.5 K shows that at about 2 T, Eu is oriented perpendicular to the external field (EuS magnetization crosses the zero). At remanence, one expects that Co lies in the film

plane, about 15° away from the beam (field) direction and Eu is antiparallel to it, so practically XMCD probes the full magnetic moment of Eu. It is interesting to notice (see inset of Fig. 4) that at 25 K, which is 50% higher than bulk T_C of EuS, the Eu XMCD signal is very large, only about 2 times smaller at remanence than at 2.5 K while it shows no saturation at 17 T. For comparison, we plot in Fig. 4 the R.T. XMCD signal. This signal is about 14 times smaller than the maximum signal at 2.5 K. If Eu[++] has a moment of 7 μ_B/ion at 2.5 K, then the R.T. moment should be 0.5 μ_B/ion, i.e., comparable to the moment of ferromagnetic Ni!

In Fig. 4, we also plot the maximum XMCD spectrum for sample A. The signal at 5 T has practically the same absolute value as the signal at remanence showing that the magnetization is close to saturation. This can be seen in the inset where the Eu magnetization curve is plotted. Note that this is an inverted hysteresis curve[26], i.e., remanence is negative because at remanence Eu is antiferromagnetically oriented to Co. At 2.5 K, the XMCD signal of Eu in sample A is larger than that of sample B because, as aforementioned, sample A has a larger amount of divalent Europium than sample B.

Discussion

Our results reveal considerable spin polarization of Eu in EuS layers due to proximity to Co layers at room temperature. The key parameter with respect to previous unsuccessful efforts in literature[7-9], was the decrease of the EuS layers by a factor of about 2, i.e. to 2 nm. Figure 4 shows that this decrease results in duplication of the Eu spin-polarization at room temperature. One could state that the Eu signal is due to the formation of an extended alloy between Co and EuS at the interface. However, Co and EuS are immiscible, since they do not follow the Hume-Rothery rules. In case of immiscible constituents, see for example Au and Co, it is not only that an extended interface alloy is not favored, but the interfaces were found to be abrupt at the monolayer limit in multilayers grown via e-beam evaporation[27,28]. Especially for the system Co and EuS, Ref. 6 reports immediate phase separation between Co and EuS in samples prepared by coevaporation even at 288 K. Successive works on Co/EuS and Fe/EuS confirmed the absence of interdiffusion[9,29].

One could understand our result by recalling the pioneer works for the increase of the critical temperature of CoF_2 in proximity to FeF_2 in superlattices[30]. Using a simple mean field theory Carriço and Camley succeeded in explaining the increase by considering interface coupling without any alloying or mixing[31]. Their work clearly showed that for 6 atomic layers of CoF_2 the polarization is throughout the whole CoF_2 layer[31]. The central atomic layers become less polarized only when the temperature increases to the 80% of the magnetic transition temperature of FeF_2, but they never become unpolarized. On the other hand, when the CoF_2 layer was 30 atomic layer thick, the atomic layers beyond the fourth from the interface became fully unpolarized[31]. We have to notice that in our samples 2 nm of a EuS(100) layer corresponds to about 6 atomic layers. Therefore, they should all be strongly polarized due to proximity to Co. Successive work by Jensen et al. using more elaborate calculations demonstrated the importance of spin fluctuations as an amplifier of the interlayer exchange coupling effects on the enhancement of the Curie temperature T_C of the low T_C magnetic layer when the latter approaches the two-dimensional limit[32]. It is reasonable to state that a 6 atomic layer thick EuS layer approaches this limit. Particularly, the response of EuS to either fields or coupling at high temperatures, much higher than its bulk Curie temperature, is further enhanced since Eu[++] has a J = 7/2, while most of the magnetic atoms or ions have much lower J[15]. One should also keep in mind that the metallic Co layers could induce a small amount of electrons into the conduction band of EuS. This would enhance the probability of filling in conduction-band states in EuS. This would immediately increase the exchange interaction, which is thought to be mediated by an indirect interaction between virtually excited conduction
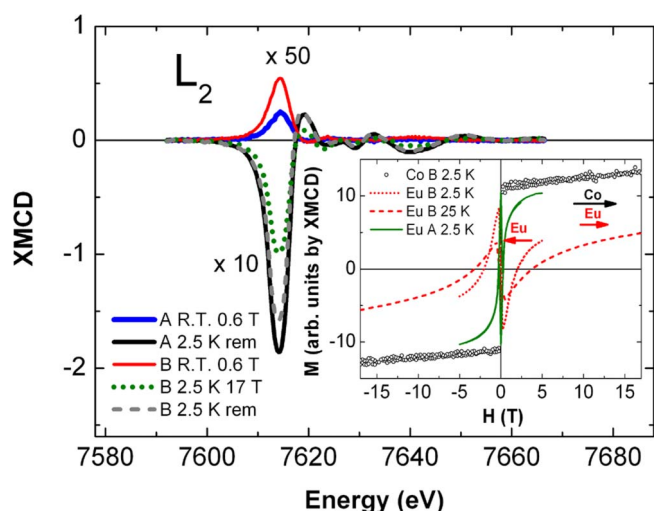

Figure 4 | Downwards: Normalized XMCD spectra at 2.5 K at the Eu L_2 edge for samples A and B. The XMCD spectra at remanence have been inverted (i.e., multiplied by a factor −1) in order to compare directly its absolute value to the spectrum at 17 T. Upwards: The spectra at R.T. have opposite direction compared to the low temperature spectra as the magnetic moment of Eu at 0.6 T is antiparallel to the Co moment. Inset: Element-specific magnetization curves for Eu and Co.

electrons and the localized Eu 4f states increasing the T_C of EuS layers[6]. For all these reasons thin EuS layers should be very susceptible to fields or direct coupling with ferromagnetic layers.

Future efforts will be focused on optimization of the growth in order to avoid the formation of trivalent Eu. This will result in another factor of 2 increase of the EuS ferromagnetism at room temperature. Finally, we have to notice that while the majority of the work on magnetic semiconductors was devoted to diluted magnetic semiconductors, recent experiments with element specificity have demonstrated that actually, despite structural perfection, there is no sign of intrinsic ferromagnetic interaction between Co substitution (dopant) ions in ZnO. The Co ions show purely paramagnetic or superparamagnetic response[33,34]. This observation enhances the value of natural ferromagnetic semiconductors at room temperature, such as EuS layers in proximity to Co layers, for future applications in the spintronics industry.

In summary, we have provided direct evidence by x-ray magnetic circular dichroism for significant spin polarization of very thin EuS layers at R.T. due to proximity to Co. For moderate magnetic fields, Eu couples antiferromagnetically to Co at R.T. where the magnetic moment of Europium is as large as that of ferromagnetic Ni. At low temperatures, even magnetic fields as large as 17 T cannot orient Eu fully parallel to Co. The strong spin polarization of the magnetic semiconductor EuS at R.T. render EuS layers thinner than about 2 nm useful in spintronic and in combinations of spintronic and optoelectronic applications.

Methods

Two Co/EuS multilayers were deposited on the native oxide of Si(100) wafers in an ultrahigh vacuum (UHV) chamber by BALZERS. The base pressure was 4×10^{-9} mbar. The deposition temperature was not higher than 100°C to avoid any layer mixing. No capping layer was used as EuS is stable in air. The deposition method for the EuS was e-beam sublimation. Here, EuS powder of high purity was placed inside a tungsten crucible and the latter one in a water-cooled e-gun body. A weak e-beam of 25 mA had to be scanned on the surface of the powder in order to get a deposition rate of 0.05 nm/sec. The material sublimes directly from the target. Co was deposited by regular e-beam evaporation with a rate of about 0.03 nm/sec. The layering quality and bilayer thickness were evaluated with the aid of the XRR technique. The XRR measurements were carried out using a Bruker Discover D8 reflectometer, equipped with a Göbel mirror, using CuKα radiation (λ = 1.5418 Å). Diffraction experiments by x-rays and electron microscopy showed *hcp* Co and EuS {100} texture.

The XMCD experiments were performed at the ID12 beamline of the European Synchrotron Radiation Facility (E.S.R.F.) in Grenoble (France) at the $L_{3,2}$ edges of Eu and the K edge of Co using a highly efficient fluorescence yield detection mode in a backscattering geometry[35]. Two magnets were dedicated for high and low field measurements: In the first one, X-ray absorption (XAS) spectra were recorded at temperatures between 2.5 and 25 K under external fields up to 17 T. In the second one, the measurements were carried out at R.T. with the help of an electromagnet working up to 0.65 T. The experiments were performed at grazing incidence (beam incidence 15° with respect to the film plane). The progress in third generation synchrotron radiation facilities has made possible the detection of the Eu XMCD signal at the Eu L-edges. The signal-to-noise ratio is large even for a single XMCD spectrum without statistics. To exclude any experimental artifacts, the XMCD spectra were recorded either by changing the helicity of the incoming light or by inverting the direction of the external magnetic field.

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Author contributions

S.D.P. and P.P. participated in the XMCD beam time. P.P. and P.F. led the project and did the main writing of the paper. B.L., A.S. and A.G. were responsible for UHV aspects, sample design and have grown the samples. V.K. has performed the XRR characterization. F.W. and A.R. are beam line scientists at ID12, ESRF and participated in the XMCD beam time. S.D.P. did complementary MOKE and XRD measurements of the samples. All authors carried out data analysis, participated in discussion of the results and commented on the manuscript.

Additional information

Competing financial interests: The authors declare no competing financial interests.

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