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Magnetotransport and antiferromagnetic coupling in nanocomposites EuS–Co

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Abstract

Nanocomposites $(EuS)_xCo_{100-x}$ ($x=30$, 50 and 70) were prepared by mechanical alloying the powders of EuS and cobalt. X-ray diffraction analysis indicated that the average particle size of EuS was reduced to about 10 nm after 50 h of milling. These EuS nanoparticles were finely mixed with the metallic cobalt. Below the Curie temperature of EuS, its moments tended to couple antiferromagnetically with that of cobalt. This macroscopic ferrimagnetic behavior was best demonstrated in the magnetization versus temperature curve of $(EuS)_{30}Co_{70}$, where a rapid decrease in the magnetization below about 16 K ($T_C=16$ K for EuS) was observed in the ball milled samples. Interesting magnetotransport behaviors were observed for $(EuS)_{70}Co_{30}$. Its magnetoresistance was positive at room temperature and changed to negative (−6.3%) at 100 K. Much larger negative magnetoresistance (~−50%) was found at 20 K. These results are discussed in the context of spin fluctuation and possible spin tunneling in the system.

Keywords: Antiferromagnetic coupling; Magnetotransport; Magnetoresistance; Nanocomposites

1. Introduction

Recent study by Gambino et al. has suggested that EuS–Co, a cobalt matrix containing precipitate particles of EuS, is a macroscopic ferrimagnet [1–4]. The co-evaporated EuS–Co film consists of ferromagnetic EuS nanoparticles which couple antiferromagnetically to the metallic cobalt matrix across phase boundary. The material has shown interesting magneto-optical properties. In addition, negative magnetoresistance was observed at and below room temperature in the system which was attributed to the spin dependent scattering at the EuS/Co interface, similar to the GMR effect. The Curie temperature of EuS was increased by a factor of 3, which has been interpreted as an exchange coupling induced effect. More recently, Rücker et al. have studied the epitaxial EuS films and Fe/EuS bilayers [5,6]. Similar enhancement of the Curie temperature of epitaxial EuS(100) films was found but attributed to the additional charge carriers introduced due to growth dislocations. The films also exhibited a peak in resistivity at the Curie temperature, which could be reduced by an applied magnetic field. In the vicinity of the peak, large negative magnetoresistance was observed. The transport phenomena were consistent with previous studies [7,8] and explained in terms of suppression of spin fluctuation by the applied magnetic fields [9,10]. In addition, antiferromagnetic interlayer coupling was observed in MBE-grown epitaxial Fe/EuS(100) bilayers [5]. We have reported the antiferromagnetic exchange coupling in mechanically milled $(EuS)_{30}Co_{70}$ below the Curie temperature of EuS [11]. The coupling strength was found to increase with decreasing particle size of EuS. This was attributed to the increased presence of EuS in the proximity of the interface between EuS and cobalt that may strongly influence the short range exchange coupling. EuS–Co is a system of great interest because it involves the magnetic interactions between two ferromagnetic materials on a nanoscale, which has not been well understood. In addition, the large negative magnetoresistance found in this system warrants further investigation. In this paper we report the magnetic and magnetotransport properties of nanocomposites $(EuS)_xCo_{100-x}$ ($x=30$, 50 and 70).

2. Experimental

Nanocomposites $(EuS)_xCo_{100-x}$ ($x=30$, 50 and 70) were prepared by mechanical alloying the powders of EuS and cobalt in a Spex-8000 mill/mixer. EuS and cobalt powders were sealed in a hardened steel vial along with hardened steel balls under helium atmosphere. The weight ratio between the balls and powders was about 4:1. X-ray

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diffraction was conducted on a Scintag powder diffractometer. The diffraction patterns of the samples after 50 h of mechanical milling could be indexed as those of cobalt and EuS. Diffraction peak analysis indicated that the average particle sizes of EuS decreased with milling time and was reduced to about 10 nm after 50 h of milling [11]. These EuS nanoparticles were finely mixed with the metallic cobalt.

Magnetization of $(EuS)_xCo_{100-x}$ was measured as a function of temperature and applied field using a Quantum Design MPMS-5S SQUID system. Resistance was also measured as a function of temperature and field on cold-pressed samples using a four point configuration.

3. Results and discussion

Shown in Fig. 1 are the magnetization *M* versus temperature *T* plots of the three milled $(EuS)_{30}Co_{70}$ samples measured in a magnetic field of 50 Oe. Also shown is that of a $(EuS)_{30}Co_{70}$ sample undergone no mechanical milling. It was hand ground for 10 min. As seen in Fig. 1, for the hand-ground sample, *M* increased suddenly as the temperature was lowered to about 16 K, which coincides with the onset of ferromagnetic ordering of EuS. A completely different behavior was observed for the mechanically milled samples. A rapid decrease in *M* below about 16 K was a common scheme among the ball milled samples. This decrease in *M* is a clear indication that the magnetization of the EuS particles align antiparallel to the magnetization of cobalt matrix when EuS is in the ferromagnetic state. Similar behavior has been observed in epitaxial Fe/EuS (100) bilayers [5]. The reduction of M increased with milling time indicating the increased strength of the antiferromagnetic coupling between the cobalt and EuS. The coupling strength was found to correlate with the particle sizes [11]. Because of the smaller sizes of the EuS particles, a larger fraction of the EuS is in close proximity to the interface and thus influenced by the exchange coupling between cobalt and EuS.

While the ordering temperatures of EuS in our ball milled $(EuS)_{30}Co_{70}$ samples remained near the Curie temperature of bulk EuS ($<$20 K), that of Gambino at al. prepared by evaporation was reported to be 3–4 times higher [1]. The ball milled samples showed no visible change of T_c as the exchange coupling was increased through longer periods of milling. This indicated that there may be no direct correlation between the T_c increase and exchange coupling between EuS and cobalt as suggested in [1]. The increase in the Curie temperature of EuS found in [1] might have been caused by other factors, for example, carrier concentration change [7]. The study of epitaxial EuS films, where T_c increase was observed in the absence of exchange coupling, supported such a mechanism [6].

Fig. 2 shows the magnetization *M* versus temperature *T* plot of $(EuS)_{70}Co_{30}$ ball milled for 50 h. The measurement was again done in an applied magnetic field of 50 Oe. An obvious feature of the EuS–Co nanocomposite at this high EuS composition was that EuS and cobalt were essentially decoupled. A possible explanation for this decoupling is the concentration of EuS had far exceeded the percolation limit [12] such that EuS formed connected networks. Although cobalt at this concentration should no longer be networked, its particle size were probably too large to have substantial coupling with the EuS. Optical microscopic examination of $(EuS)_{70}Co_{30}$ supported such claim. The magnetization of $(EuS)_{70}Co_{30}$ below the Curie temperature of EuS was the sum of two terms, contributed independently from EuS and cobalt.

Resistivity study indicated that $(EuS)_{30}Co_{70}$ and $(EuS)_{50}Co_{50}$ were metallic. Fig. 3 shows the temperature dependence of resistance of $(EuS)_{50}Co_{50}$ ball milled for

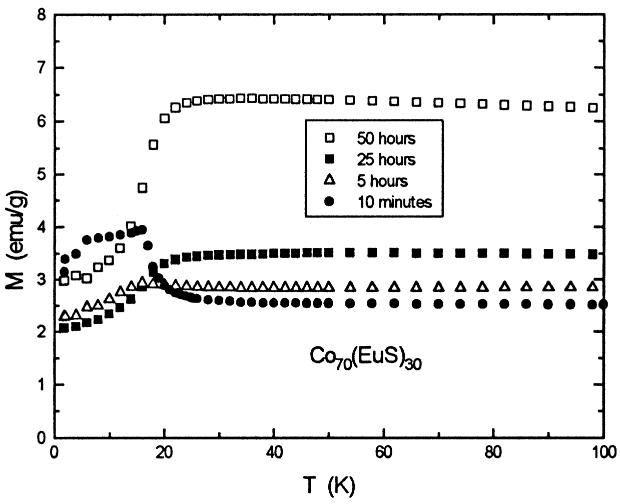

Fig. 1. Magnetization of $(EuS)_{30}Co_{70}$ after 10 min, and 5, 25 and 50 h of milling.

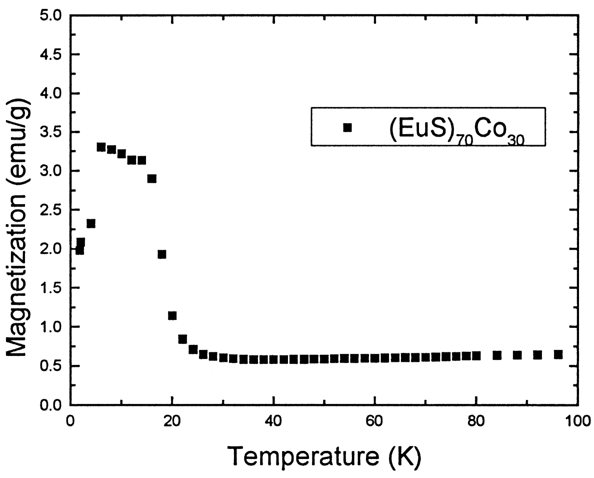

Fig. 2. Magnetization *M* versus temperature *T* plot of $(EuS)_{70}Co_{30}$ ball milled for 50 h.

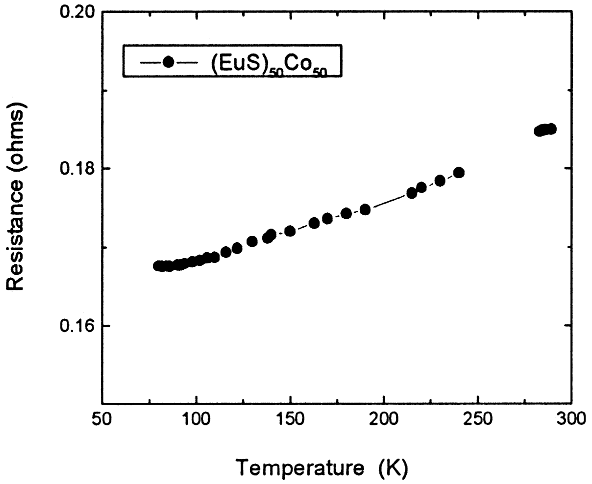

Fig. 3. Temperature dependence of the resistance of (EuS)$_{50}$Co$_{50}$.

50 h. It exhibited a typical metallic behavior, which was mainly attributed to the cobalt.

Interesting transport behaviors were observed for (EuS)$_{70}$Co$_{30}$. Its resistivity value was close to that of a semiconductor. As temperature was decreased, the resistivity initially increased, reached a broad maximum at about 220 K, and then decreased again (Fig. 4). Similar temperature dependence of resistivity has been observed in EuO, which was due to carrier density variation as a function of temperature [13]. A model consisting of a donor-trap level, possibly caused by an oxygen vacancy, which crosses the conduction band edge from above with increasing temperature could account for much of the observed behavior in EuO. What have been seen in our (EuS)$_{70}$Co$_{30}$ resembled that of EuO and, possibly, the same mechanism gave rise to both. Obviously, the reason for the absence of metallic conductivity in (EuS)$_{70}$Co$_{30}$ was that the concentration of cobalt was too low to form connected networks.

Below 50 K, two types of resistivity were observed. One (EuS)$_{70}$Co$_{30}$ (Sample A) showed a maximum at ~16 K (see Fig. 4, inset), and the other (Sample B) showed a minimum at ~25 K (see Fig. 4). Works are currently underway to examine in detail the discrepancy between the two. However, both behaviors have been observed in ferromagnetic semiconducting europium chalcogenides EuS and EuO. The peak near the Curie temperature of EuO or EuS are believed to be associated with critical scattering due to spin fluctuation [6–10]. For EuO there seems to be a general agreement between the experimental data and the theory based on the spin fluctuation. For EuS, on the other hand, limited data also suggests a possible metal insulator transition over the temperature region of the resistivity peak [7]. More study is needed to understand the exact origin of the resistivity peak in EuS, at the same time this anomaly resembles some of the features of the colossal magnetoresistive (CMR) perovskites. As will be shown next, large negative magnetoresistance was detected in (EuS)$_{70}$Co$_{30}$. A minimum in resistivity at 15 K has been observed in EuO, which was ascribed to an impurity hopping mechanism [14] and activation of carriers from a rare earth impurity into the conduction band [13]. Considering the similar crystal and electronic structures between EuO and EuS, such mechanisms are plausible for the minimum near 25 K in one of our (EuS)$_{70}$Co$_{30}$ samples (Sample B).

The magnetoresistance of (EuS)$_{70}$Co$_{30}$ was positive at room temperature but changed to negative (−6.3%) at 100 K. Much larger negative magnetoresistance (~−50%) was found at 20 K. These results are shown in Fig. 5. The magnetoresistance data of (EuS)$_{70}$Co$_{30}$ above the Curie temperature seemed to indicate that much of the negative

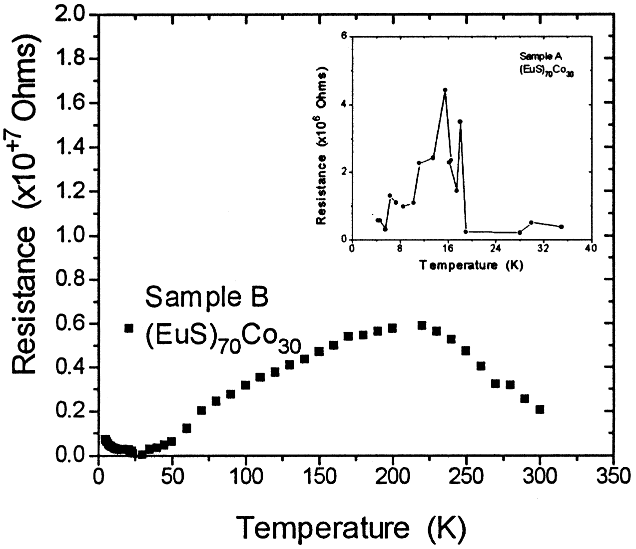

Fig. 4. Temperature dependence of the resistance of (EuS)$_{70}$Co$_{30}$. Sample A showed a maximum at ~16 K (inset), and Sample B showed a minimum at ~25 K.

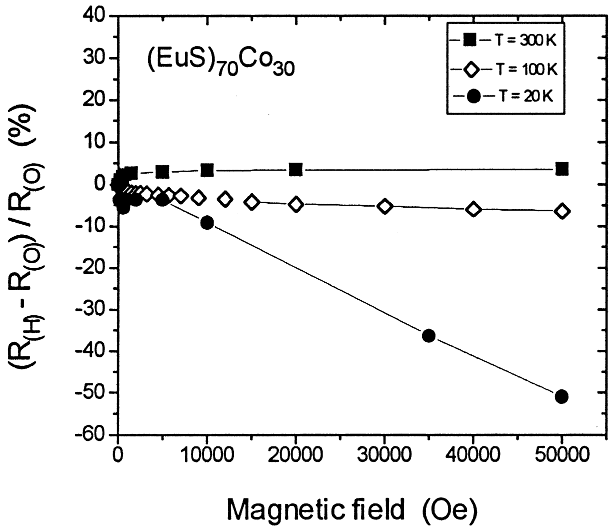

Fig. 5. Magnetoresistance of (EuS)$_{70}$Co$_{30}$ at 20, 100 and 300 K, respectively.

magnetoresistance was because of the suppression of the spin fluctuation by the applied magnetic fields. As the spin fluctuation was quenched by the fields, the resistivity peak was smeared off, which led to a larger reduction in resistivity near the peak region (e.g., at 20 K) than 100 K. At room temperature, the magnetoresistance due to spin fluctuation alone was minimal and the positive magnetoresistance observed was that intrinsic to a semiconductor.

In Fig. 6 the magnetoresistance of $(EuS)_{70}Co_{30}$ at 12 K, below the Curie temperature, is shown. The behavior was typical of a ferromagnet. The initial increase of resistivity at low fields was due to spontaneous resistivity anisotropy. After the saturation of the magnetization, the decrease in resistivity upon further field increase was because of the reduction of spin disorder resistivity [15].

It should be mentioned that, above the Curie temperature of EuS, the negative magnetoresistance of $(EuS)_{70}Co_{30}$ might also arise from the spin polarized tunneling. At this composition, one can consider that cobalt particles were imbedded in EuS matrix. A possible pathway for the electrons was to tunnel through the semiconducting barrier EuS between two magnetic metal particles. Negative magnetoresistance has been observed and understood in terms of spin polarized tunneling in both layered and granular systems where electrons tunnel between magnetic metals through an insulator barrier [16–18]. There is currently much interests in spin tunneling and GMR materials and the possible connection between the spin tunneling and the present system should be of significance. Experimental evidence supporting such speculation was that no negative magnetoresistance below the Curie temperature of EuS was observed in $(EuS)_{70}Co_{30}$. On the contrary, negative magnetoresistance has been found on both sides of the Curie temperature for pure EuS and EuO, where spin fluctuation gave rise to the negative magnetoresistance.

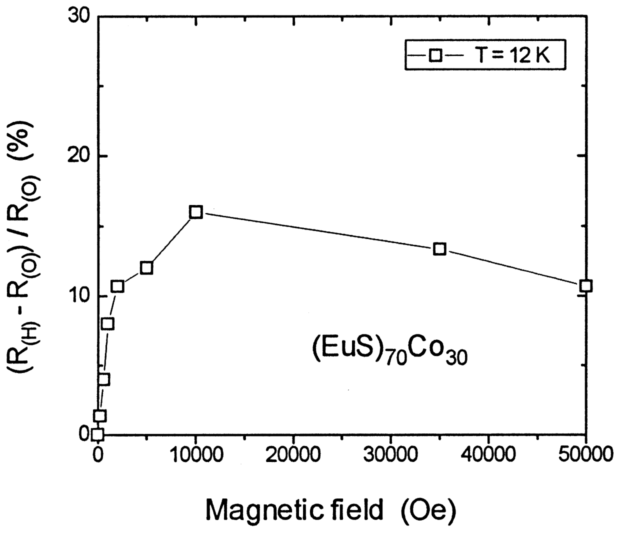

Fig. 6. Magnetoresistance of $(EuS)_{70}Co_{30}$ at 12 K, below the Curie temperature of EuS.

4. Conclusions

Nanocomposites $(EuS)_xCo_{100-x}$ ($x=30$, 50 and 70) was prepared by mechanical alloying the powders of EuS and cobalt. For $x=30$, EuS nanoparticles were uniformly distributed in the cobalt matrix. Below the Curie temperature of EuS, their moments coupled antiferromagnetically with that of cobalt. This antiferromagnetic exchange coupling gave rise to the novel behaviors shown in its magnetization versus temperature curve. The composites exhibited metallic conductivity up to $x=50$. For $x=70$, the composite showed a temperature dependence of the resistivity that could be explained in terms of carrier concentration change of semiconducting EuS. At this composition, a large negative magnetoresistance was found (−50% at 20 K), which decreased with increasing temperature. It was switched to positive at room temperature. The negative magnetoresistance above the Curie temperature of EuS resulted most likely from the critical scattering due to spin fluctuation. But the possibility that it was associated with spin polarized tunneling could not be ruled out.

Acknowledgements

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