Magnetic properties of (Eu,Gd)Te semiconductor layers

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In (Eu,Gd)Te semiconductor alloys, a well known antiferromagnetic semiconductor compound EuTe is transformed into an n-type ferromagnetic alloy. This effect is driven by the RKKY interaction via conducting electrons, created due to the substitution of Gd^{3+} for Eu^{2+} ions. It is expected that due to the high degree of electron spin polarization, (Eu,Gd)Te can be exploited in new semiconductor spintronic heterostructures as a model injector of spin-polarized carriers. (Eu,Gd)Te monocrystalline layers with Gd content up to 5 at. % were grown by MBE on BaF_2 (111) substrates with either PbTe or EuTe buffer layers. Measurements of magnetic susceptibility and magnetization revealed that the ferromagnetic transition with the Curie temperature of $T_C = 11$–15 K is observed in (Eu,Gd)Te layers with n-type metallic conductivity. The analysis of the magnetization of (Eu,Gd)Te was carried out in a broad range of magnetic fields applied along various crystal directions, both in- and out of the layer plane. It revealed, in particular, that a rapid low-field ferromagnetic response of the (Eu,Gd)Te layer is followed by a paramagnetic-like further increase towards a full saturation.

Key words: *spintronics; ferromagnetic semiconductor; rare earth compound*

1. Introduction

Europium telluride belongs to a well know family of magnetic semiconductors and is a model type II antiferromagnetic material. This material offers the unique possibility to transform it from an insulating antiferromagnetic to an n-type ferromagnetic by substituting Gd^{3+} ions for Eu^{2+} in the crystal matrix. The mechanism responsible for such a transformation is attributed to Ruderman–Kittel–Kasuya–Yosida interactions, i.e. the coupling between strongly localized Eu $S = 7/2$ spins mediated by quasi-free carriers. In early studies of bulk materials, it was shown [1–4] that for concentrations of Gd up to 60 at. %, (Eu,Gd)Te crystals exhibit a ferromagnetic transition, whereas for higher concentrations of Gd the type of magnetism again changes to antiferromag-

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netic (resulting from the oscillating behaviour of RKKY interactions). According to the above-described mechanism, all europium-gadolinium chalcogenides with Gd contents below about 60 at. % were found to be ferromagnetic semiconductors with the maximum Curie temperature $T_C = 150$ K for oxides and about $T_C = 10$ K for tellurides [1–4]. The terminal alloys (gadolinium chalcogenides) are antiferromagnetic compounds with metallic-type conductivity. In the n-type ferromagnetic state, (Eu,Gd)Te exhibits high electron-spin polarization related to the large splitting of 5d6s conduction band states. This feature, together with the expected epitaxial compatibility of (Eu,Gd)Te with well-known nonmagnetic semiconductor materials such as PbTe or CdTe, makes (Eu,Gd)Te an intriguing key-element of new all-semiconductor spin injection spintronic heterostructures. In this work, we experimentally study the magnetic properties of epitaxial layers of (Eu,Gd)Te in which both metallic and insulating electrical properties are observed, depending on Gd content and the crystal stoichiometry of the alloy.

2. Growth and characterization of layers

The (Eu,Gd)Te layers were grown in a home-built MBE system equipped with effusions cells for Eu, Gd, Te_2, and PbTe solid sources. Either a EuTe layer or a EuTe/PbTe bilayer, deposited on a freshly cleaved (111) surface of BaF_2 monocrystals at about 270 °C, was used as a buffer. Next, a (Eu,Gd)Te layer from 0.25 up to 1 µm thick was grown. Reflection high-energy electron diffraction (RHEED) was used to monitor the growth process in situ, showing well-defined streaky patterns indicating the two-dimensional mode of growth. A more detailed description of the MBE growth process for (Eu,Gd)Te layers is given in Ref. [5]. Structural studies of epilayers were carried out using standard X-ray diffraction (XRD). The θ–2θ diffraction spectra clearly revealed the peaks due to the BaF_2 substrate, the buffer layer, and (Eu,Gd)Te layer with lattice parameters depending on Gd content. All the layers are monocrystalline (rock salt structure) with the [111] growth direction. FWHM parameters of the rocking curves in the range 200–400 arcsec confirmed the good crystalline quality of our samples. Additionally, epilayers were examined by atomic force microscopy (AFM). The root mean square parameter describing the roughness of the layer surface was 0.6 nm (corresponding to two atomic monolayers) over a 10×10 µm^2 area for the best layers. The concentration of gadolinium in the layers was obtained from energy dispersive X-ray fluorescence and electron micro-probe analysis. (Eu,Gd)Te layers with a maximum content of Gd up to 5 at. % were obtained.

3. Experimental

AC magnetic susceptibility (χ) measurements were applied to investigate the influence of quasi-free carriers on the type of magnetic ordering in (Eu,Gd)Te layers. The measurements were carried out in the temperature range $T = 4.5$–80 K using

a LakeShore susceptometer and applying an AC magnetic field of 5 Oe at the frequency of 625 Hz. We obtained two dramatically different $\chi(T)$ curves, depending on the type of conductivity (Fig. 1).
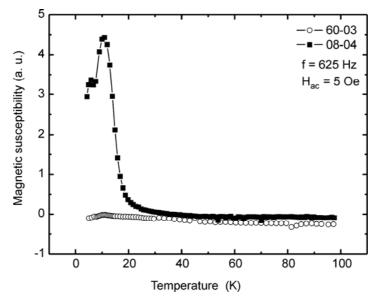

Fig. 1. AC magnetic susceptibility vs. temperature: insulating
antiferromagnetic (open dots) and n-type ferromagnetic (squares) layer

For all n-type (Eu,Gd)Te layers, a ferromagnetic transition was observed (squares) at T_C = 11–15 K, while insulating (Eu,Gd)Te layers exhibited an antiferromagnetic transition (open dots) at $T_N \approx 10$ K. In the latter case, in spite of incorporating Gd into the EuTe matrix (therefore supplying it with carriers), we destroyed n-type conductivity using excess Te in the molecular beam flux, resulting in an insulating (Eu,Gd)Te material similar to undoped EuTe. These experimental observations give strong evidence of carrier-induced ferromagnetic ordering in (Eu,Gd)Te alloys. The magnetic effects discussed above were also reflected in electron transport studies. The Hall effect measurements, carried out in the temperature range 4.2–300 K, revealed n-type conductivity with a very high concentration of quasi-free carriers $n \approx 10^{20}$ cm^{-3}. Moreover, the resistivity dependence on temperature demonstrates behaviour correlated with the magnetic transition. Lowering T_C results in an increase of resistance due to the scattering on spin moments. Over 90% negative magnetoresistance was observed at magnetic fields of about 0.6 T. The ferromagnetic properties of n-(Eu,Gd)Te were also studied by SQUID magnetometry. Measurements of magnetization hysteresis loops were carried out in various crystal directions, both in-plane and out of plane (Fig. 2). In (Eu,Gd)Te layers, the in-plane magnetocrystalline anisotropy is very weak, with an estimated anisotropy field below 2 mT. The dominant anisotropy effect is the strong ($H_A \approx 1$ T) shape anisotropy expected in a ferromagnetic layer.
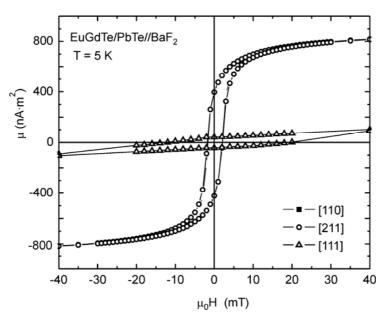
Fig. 2. Hysteresis loops for various crystalline directions:
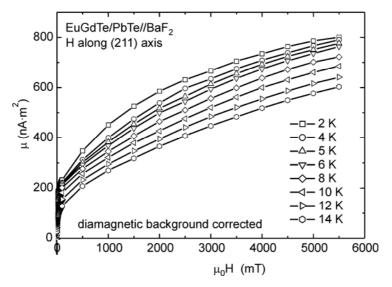out-of-plane [111] as well as in-plane ([110] and [211])

Fig. 3. High-field magnetization of n-(Eu,Gd)Te layers (SQUID measurements)

SQUID high-field measurements of magnetization for n-(Eu,Gd)Te (see Fig. 3) showed that the low-field rapid ferromagnetic response is followed by a much slower approach to full magnetic saturation, still not reached at fields of 5.5 T. Recent MOKE magnetometry studies of high-field (up to 20 T) magnetization of n-(Eu,Gd)Te layers showed full saturation (at $T = 5$ K) for fields above 10 T. Although this behaviour is not yet fully un-

derstood, we consider in particular the competition between ferromagnetic and anti-ferromagnetic exchange interactions when discussing this experimental finding. We also consider possible electronic separation effects, resulting in the co-existence of n-type ferromagnetic and insulating antiferromagnetic regions in the layer.

4. Conclusions

Heterostructures incorporating (Eu,Gd)Te semiconductor ferromagnetic layers were grown on BaF_2 (111) substrates exploiting the MBE technique. Structural characterization performed by RHEED, XRD, and AFM methods revealed good surface morphology and crystal quality of the layers. The Hall effect and resistivity measurements showed the dependence of electrical properties of (Eu,Gd)Te on stoichiometry (controlled by tellurium content), resulting in either a metallic n-type or insulating material. The ferromagnetic transition with the Curie temperature of T_C = 11–15 K was observed only in metallic n-(Eu,Gd)Te layers.

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