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(Eu,Gd)Te — MBE Growth and Characterization

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Monocrystalline thin layers of (Eu,Gd)Te, n-type ferromagnetic semiconductor, were grown by molecular beam epitaxy technique on BaF_2 (111) substrates. Reflection high-energy electron diffraction, X-ray diffraction, and atomic force microscopy characterization proved epitaxial mode of growth and high crystal quality of the layers. Magnetic susceptibility and magnetic resonance measurements showed that in (Eu,Gd)Te layers ferromagnetic transition takes place at about 13 K. Electrical characterization carried out by the Hall effect and resistivity measurements revealed very high electron concentration of 10^{20} cm^{-3} and sharp maximum of resistivity at transition temperature.

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1. Introduction

Development of new bipolar semiconductor spintronic heterostructures requires suitable, epitaxial quality n-type ferromagnetic semiconductor layers. Unique possibilities are offered here by a model material system of the family of europium-gadolinium chalcogenides [1–3]. The bulk crystals of the substitutional solid solutions of $Eu_{1-x}Gd_xTe$, $Eu_{1-x}Gd_xSe$, $Eu_{1-x}Gd_xS$, and $Eu_{1-x}Gd_xO$ crystallize (in the entire solubility range, $0 \le x \le 1$) in the rock salt structure. These materials show n-type metallic conductivity with a very high electron concentration of $n = 10^{20}$–10^{22} cm^{-3} resulting from the substitution of Gd^{3+} for Eu^{2+} ions in the cation sublattice. For Gd content $x < 0.5$, these materials exhibit ferromagnetic transition driven by the Ruderman–Kittel–Kasuya–Yoshida

(RKKY) mechanism via the spin polarization of conduction electrons with the maximal Curie temperature (for $x \approx 0.1$) varying between $T_{\mathrm{C}} = 10$ K for tellurides up to $T_{\mathrm{C}} = 150$ K for oxide materials [1–6]. For higher content of Gd ($x > 0.5$), the extremely high concentration of quasi-free electrons results in the increasing importance of the well known effect of oscillating (ferromagnetic/antiferromagnetic) sign of the RKKY interaction, eventually leading to the destruction of the ferromagnetic order. The terminal ($x = 1$) compounds GdTe, GdSe, GdS, and GdO are metallic RKKY antiferromagnets with the Néel temperature $T_{\mathrm{N}} = 50$ K for GdTe [1–6].

In this work we demonstrate the growth by molecular beam epitaxy (MBE) technique of (Eu,Gd)Te layers on BaF_2 (111) substrates with EuTe–PbTe buffer bilayer as well as discuss the results of their structural, magnetic, and electrical characterization. We exploit here important technological features of (Eu,Gd)Te, i.e., very good match of lattice parameters and thermal expansion coefficients of (Eu,Gd)Te, PbTe, and some semimagnetic (diluted magnetic) IV–VI semiconductors with Eu, as well as the availability of good quality monocrystalline BaF_2 substrates.

2. Growth and structural characterization

The (Eu,Gd)Te layers were grown by MBE technique on freshly cleaved (111) surface of BaF_2 monocrystals using effusion cells for Eu, Gd, and Te_2. The substrate temperature was about 270°C. EuTe–PbTe bilayer was usually used as a buffer. The thickness of (Eu,Gd)Te layer typically was about 0.5–1 μm whereas the thickness of the buffer layer was about 0.1 μm. The maximal content of Gd in good quality (Eu,Gd)Te layers was 5 at.% as checked by electron microprobe and energy dispersive X-ray fluorescence analysis, while the typical content of Gd in a set of about 20 samples grown for this study was approximately 1 at.%.

The growth of the (Eu,Gd)Te layers and the EuTe–PbTe buffer layer was monitored *in situ* by reflection high energy electron diffraction (RHEED) technique revealing well defined streaky pattern characteristic of two-dimensional mode of growth (see Fig. 1a). In some cases very clear oscillations of the intensity of specular spot of RHEED pattern were observed (see Fig. 1b) allowing for the precise determination of the rate of growth, found to be about 0.1 ML/s, where for (Eu,Gd)Te crystal lattice in [111] growth direction 1 monolayer (ML) is 0.37 nm. Post-growth, the crystal quality of the layers was examined by standard X-ray diffraction (XRD) method. An example of such an analysis performed for the (Eu,Gd)Te layer with high Gd content is presented in Fig. 2. The relatively small width of the X-ray rocking curve (300–600 arcsec) observed in (Eu,Gd)Te layers proves their good crystalline quality and allows for clear separation of diffraction peaks originating from BaF_2 substrate, PbTe and EuTe buffer layers and the (Eu,Gd)Te layer. The doublet (K_α, K_β line of Cu) spectral character of the

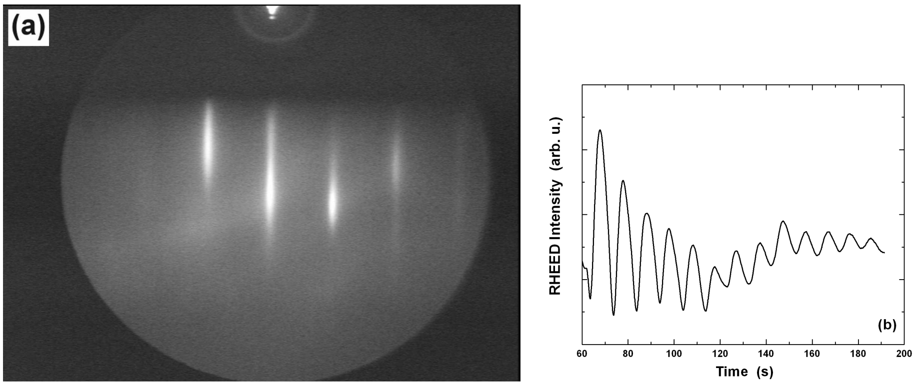

Fig. 1. The RHEED *in situ* characterization of the growth process of (Eu,Gd)Te layer. The well defined streaky diffraction pattern (a) reveals two-dimensional mode of epitaxial growth, whereas the observation of the oscillations of the intensity of specular spot of RHEED diffraction pattern (b) allows precise determination of the growth rate.

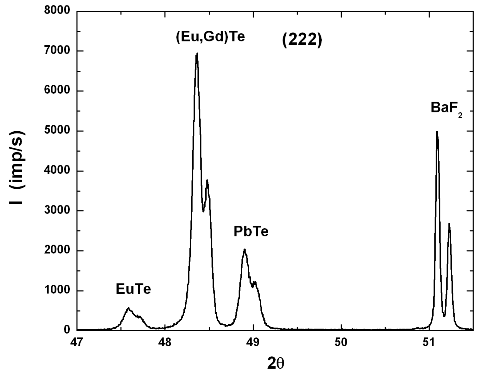

Fig. 2. The standard X-ray θ–2θ diffractogram of (Eu,Gd)Te layer grown on BaF_2 (111) substrate with PbTe/EuTe bilayer buffer.

X-ray source is clearly reflected in the (Eu,Gd)Te–EuTe–PbTe/BaF_2 (111) diffraction pattern giving another evidence for high crystal quality of the layers. The surface morphology of the layers was studied by atomic force microscopy (AFM). AFM investigations were performed at room temperature in the air revealing good smoothness of the surface of many (Eu,Gd)Te layers with the root mean square (RMS) roughness parameter equal to 0.6 nm (for the analysed area of 10×10 μm^2). An example of such an analysis is presented in Fig. 3.

The growth of (Eu,Gd)Te and EuTe layers was performed under various technological conditions controlled in the MBE chamber. In particular, it concerned the substrate temperature and the ratio of the molecular fluxes of Eu and Te_2. Particularly important for the growth of (Eu,Gd)Te layers turned out to be the control of their stoichiometry. Good quality n-type metallic layers exhibiting

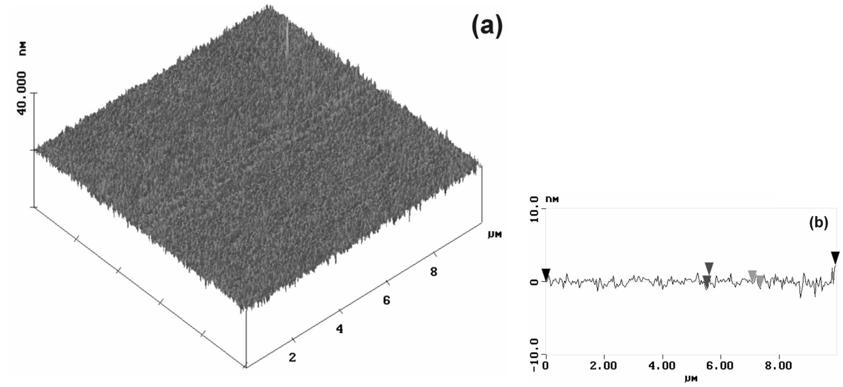

Fig. 3. The atomic force microscopy characterization of the surface of (Eu,Gd)Te layer. Part (a) presents the area scan over 10 × 10 μm^2, whereas part (b) shows the selected linear scan indicating almost atomic surface flatness over microscopic distance of 10 μm.

ferromagnetic transition were obtained only during the growth performed under conditions close to stoichiometric, whereas the growth of (Eu,Gd)Te layers with large excess Te_2 flux resulted in insulating and paramagnetic layers. It is likely that the other (than EuTe and GdTe) telluride compounds of Gd and Eu can be formed under such conditions. For example in Eu_2Te_3 and Gd_2Te_3, the Eu and Gd ions are in 3+ charge state and the substitution of Gd for Eu produces no free electrons. The presence of certain concentration of Eu^{3+} ions in (Eu,Gd)Te layers was recently observed in photoemission spectroscopy studies of the surface of these layers [7]. Both surface oxidation and deviations from stoichiometric composition are the possible driving mechanisms for the change of Eu valence from 2+ (expected in EuTe) to 3+ one.

3. Magnetic and electrical characterization

For the magnetic characterization of the layers of (Eu,Gd)Te, the magnetic susceptibility, χ, was measured in the temperature range T = 4.5–80 K using LakeShore susceptometer and applying the ac magnetic field of 5 Oe at frequency of 625 Hz. Figure 4a presents the $\chi(T)$ dependence observed in all (Eu,Gd)Te layers exhibiting n-type conductivity. The sharp increase in the susceptibility and the maximum on the $\chi(T)$ plot clearly indicate the ferromagnetic transition at T_C = 11–13 K (slightly differing in various layers). The diamagnetic, practically temperature independent, background observed on $\chi(T)$ dependence at higher temperatures is due to the contribution of BaF_2 substrate. In (Eu,Gd)Te–EuTe–PbTe multilayers the magnetic contributions of antiferromagnetic layer of EuTe and diamagnetic layer of PbTe are negligible. It is clearly observed in (Eu,Gd)Te–EuTe/BaF_2 layers grown under high excess Te conditions. These layers are insulating and show no ferromagnetic transition. In these layers the magnetic suscep-

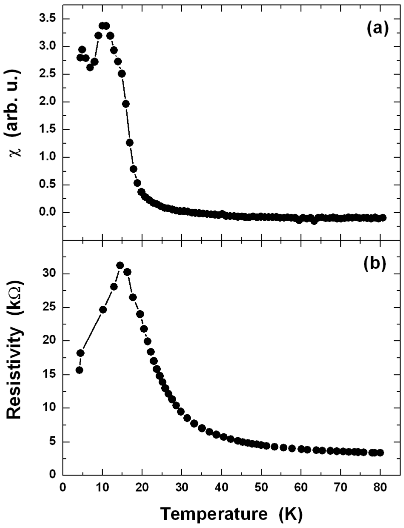

Fig. 4. The temperature dependence of (a) the ac magnetic susceptibility and (b) the electrical resistance of (Eu,Gd)Te–EuTe/BaF_2 layer exhibiting ferromagnetic transition at about 13 K.

tibility measurements show only a small cusp at $T \approx 10$ K corresponding to the Néel temperature of antiferromagnetic transition in EuTe and (Eu,Gd)Te insulating layers.

The layers of (Eu,Gd)Te were also studied by electron magnetic resonance technique employing X-band Bruker spectrometer with magnetic field 3.4 kOe corresponding to the g-factor 2.0. The measurements confirmed the ferromagnetic transition in (Eu,Gd)Te layers as evidenced by, characteristic of ferromagnetic resonance, angular and temperature dependence of the resonant field. At $T = 2\ \mathrm{K} < T_\mathrm{C}$, the maximal experimentally observed resonance field is 8.5 kOe for external magnetic field applied normally to the plane of the layer. This is due to, well known in ferromagnetic thin layers, effect of shape anisotropy brought about by dipolar interactions. The shape anisotropy strongly energetically favours in-plane location of the magnetization vector of (Eu,Gd)Te layers. The experimentally observed magnitude of the shape anisotropy corresponds (at $T = 2$ K) to about 50% of the magnitude of local magnetization expected for fully magnetically saturated system of 7 μ_B (Bohr magneton) spins in (Eu,Gd)Te. This experimental finding indicates that either the magnetic order in (Eu,Gd)Te is of non-collinear type or the microscopic (electrical or magnetic) homogeneity of the layers is yet not perfect.

For the electrical characterization of low resistivity (Eu,Gd)Te layers, the temperature dependence of the Hall effect and resistivity was studied in the temperature range $T = 4.2$–300 K. The Hall effect measurements revealed n-type conductivity with very high concentration of quasi-free electrons $n \approx 10^{20}\ \mathrm{cm}^{-3}$.

This observation was additionally confirmed by thermoelectric power measurements. Due to the strong magnetic dipolar anisotropy observed in (Eu,Gd)Te and resulting in the in-plane orientation of magnetization vector, the Hall effect measurements performed in the standard geometry (with external magnetic field applied normally to the plane of the layer) reveal only small contribution of anomalous Hall effect. This effect is known to be proportional to the normal to the layer component of the magnetization. The ferromagnetic transition is, however, strongly reflected in other electron transport effects as evidenced by a sharp peak on the temperature dependence of resistivity (see Fig. 4b) as well as large negative magnetoresistance of about 100% at magnetic field of 10 kOe.

4. Summary

In summary, employing the technique of molecular beam epitaxy, the monocrystalline thin layers of (Eu,Gd)Te semiconductor were grown on BaF_2 (111) substrates with EuTe–PbTe buffer bilayer. The structural characterization carried out with the use of RHEED, XRD, and AFM methods proved epitaxial mode of growth and high crystal quality of the layers. The electrical properties of the layers strongly depend on the stoichiometry: both metallic n-type layers as well as insulating (compensated by excess Te) layers were obtained. Both magnetic susceptibility and magnetic resonance measurements showed that the n-type (Eu,Gd)Te layers exhibit ferromagnetic transition with the Curie temperature $T_C = 11$–13 K in various layers. The ferromagnetic transition influences also the electron transport processes as evidenced by the sharp maximum of resistivity at the transition temperature.

Acknowledgments

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