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Transport and magnetic properties of $Ge_{1-x-y}Mn_x(Eu,Yb)_yTe$ semimagnetic semiconductors

B. BRODOWSKA[1*], I. KURYLISZYN-KUDELSKA[1], M. ARCISZEWSKA[1], K. DYBKO[1],
V. DOMUKHOVSKI[1], W. DOBROWOLSKI[1], V.E. SLYNKO[2], E.I. SLYNKO[2], V.K. DUGAEV[3]

[1]Institute of Physics, Polish Academy of Sciences, al. Lotników 32/46, 02-668 Warsaw, Poland

[2]Institute for Problems of Material Science, NASU, Vilde 5, 58001 Chernovtsy, Ukraine

[3]Department of Mathematics and Applied Physics, Rzeszów University of Technology,
al. Powstańców Warszawy 6, 35-959 Rzeszów, Poland

Transport and magnetic properties of the IV–VI ferromagnetic multinary mixed compounds $Ge_{1-x}Mn_xTe$, $Ge_{1-x-y}Mn_xEu_yTe$ and $Ge_{1-x-y}Mn_xYb_yTe$ ($0.045 \le x \le 0.47$, $y < 0.04$) were investigated in magnetic fields up to 13 T and in the temperature range 1.6–300 K. The Curie temperatures of studied samples exceed 100 K. Based on the dependences of the Hall resistivity and magnetization on magnetic field, the anomalous Hall coefficients R_S were calculated, and R_S dependences on crystal composition and temperature were found.

Key words: *semimagnetic semiconductor; diluted magnetic semiconductor; anomalous Hall effect*

1. Introduction

It has been well known from early 70s that even a small admixture of MnTe in GeTe leads to the ferromagnetic behaviour of the resulting alloy [1]. The mixed compound $Ge_{1-x}Mn_xTe$ for $x < 0.18$ crystallizes in the rhombohedrally distorted NaCl structure. $Ge_{1-x}Mn_xTe$ belongs to the interesting class of materials which simultaneously reveal ferromagnetic and ferroelectric properties [2]. These features may lead to interesting applications in spintronic devices. $Ge_{1-x}Mn_xTe$ crystals are always p-type and the ferromagnetism is accounted to the Ruderman–Kittel–Kasuya–Yosida interaction [3] which proceeds via a coupling between the ion and the mobile charge carriers. The magnetic Curie temperature depends on both the Mn content and the carrier concentration, and its highest reported value does not exceed 160 K. In our recent paper, we revealed that a small admixture of rare earth ion Eu in GeMnTe substantially increased the ferromagnet–paramagnet phase transition temperature [4]. The effect is not

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[*] Corresponding author, e-mail: brodo@ifpan.edu.pl

yet fully understood, and some attempts of possible explanations may be found in [4]. In the present paper, we report results of magnetotransport and magnetic studies of the bulk crystals of $Ge_{1-x}Mn_xTe$ as well as $Ge_{1-x}Mn_xTe$ codoped with Eu and Yb. Based on the dependences of Hall resistivity and magnetization on magnetic field, the anomalous Hall coefficient R_S was calculated, and R_S dependences on crystal composition and temperature were determined.

2. Experimental

The $Ge_{1-x}Mn_xTe$, $Ge_{1-x-y}Mn_xEu_yTe$ and $Ge_{1-x-y}Mn_xYb_yTe$ bulk crystals used in this study were grown by the modified Bridgman method. The chemical compositions of the crystals were determined by the X-ray dispersive fluorescence technique. The manganese content x was found to be in the range 4.5–47 at. % while the rare earth ions content did not exceed 4 at. %. The X-ray powder diffraction studies showed that the crystals were single-phase, except the samples $Ge_{1-x-y}Mn_xTe$ – 960_12 and $Ge_{1-x-y}Mn_xYb_yTe$ – 971_12, where the traces of a second phase were visible. The sample compositions and carrier concentrations derived from the transport measurements are given in Table 1.

Table 1. Samples characteristics

Sample symbol	Composition [molar fraction]	Hole concentration [cm^{-3}]
$Ge_{1-x}Mn_xTe$ – 960_2	$x = 0.045$	1.04×10^{21}
$Ge_{1-x}Mn_xTe$ – 960_12	$x = 0.388$	2.86×10^{21}
$Ge_{1-x-y}Mn_xEu_yTe$ – 849_8	$x = 0.087, y = 0.038$	1.06×10^{21}
$Ge_{1-x-y}Mn_xEu_yTe$ – 849_16	$x = 0.073, y = 0.03$	1.01×10^{21}
$Ge_{1-x-y}Mn_xYb_yTe$ – 971_12	$x = 0.47, y = 0.004$	4.16×10^{21}

The magnetic properties were studied by use of the Lake Shore 7229 DC magnetometer/AC susceptometer system: ac magnetic susceptibility in function of temperature up to 170 K (by means of a standard mutual inductance method), and magnetization in magnetic fields B up to 9 T (by an extraction method) in the temperature range 1.5–170 K. The examples of dependences of the ac susceptibility on temperature are presented in Fig. 1. The magnetization curves vs. magnetic field for two samples are shown in Fig. 2. It may be noted that in the available range of magnetic fields we do not observe the saturation of magnetization, particularly large discrepancies between experimental values and the expected ones were observed for the samples with the highest manganese contents. For example, the theoretically calculated saturation magnetization for the sample $Ge_{1-x-y}Mn_xYb_yTe$ 971_12 (see Fig. 2) equals 68.3 emu/g, approximately three times higher than the experimental value obtained at 9 T. This phenomenon is likely due to the presence of antiferromagnetic nanoinclusions assembled due to chemical or crystallographic phase separations. These nanoclusters are below the detection limit of the conventional powder diffraction method. Nanoinclusions (of ferromagnetic or antiferromagnetic nature) were found in several semimagnetic materials [5], and it was established that their presence significantly modifies properties of the host material.

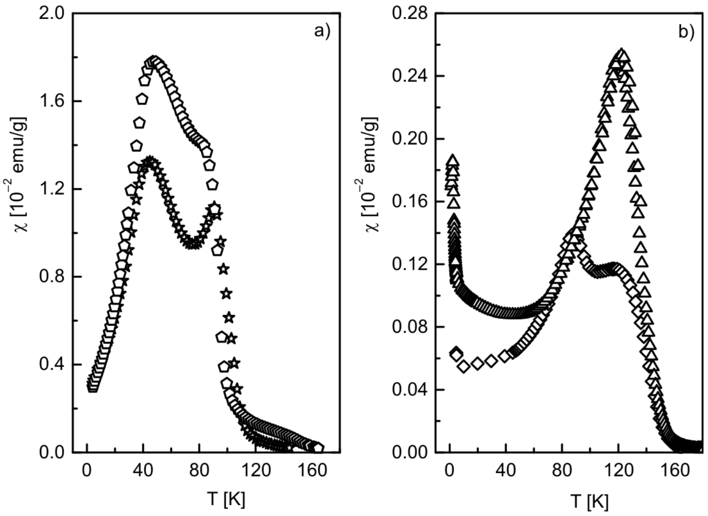

Fig. 1. Magnetic susceptibility of: a) ★ – $Ge_{1-x}Mn_xTe$, $x = 0.388$, ⬠ – $Ge_{1-x-y}Mn_xYb_yTe$, $x = 0.47$, $y = 0.004$, b) ◇ – $Ge_{1-x}Mn_xTe$, $x = 0.045$, △ – $Ge_{1-x-y}Mn_xEu_yTe$, $x = 0.087$, $y = 0.038$

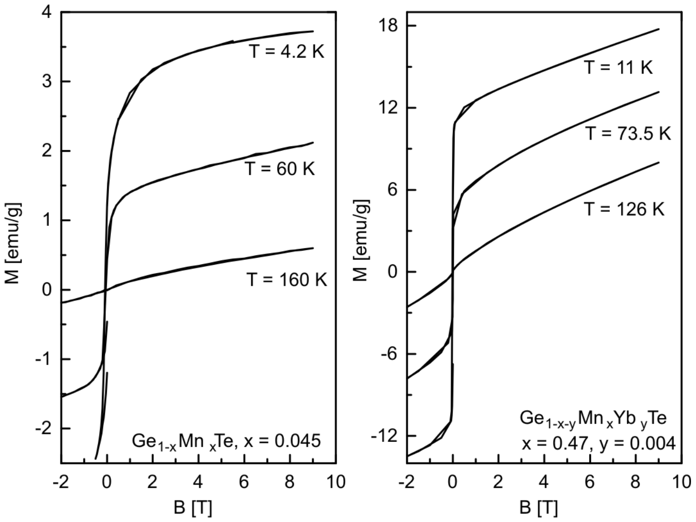

Fig. 2. Magnetization of selected samples for low (a) and high (b) manganese contents, at three temperatures below and near (above) the Curie temperature

The Hall resistivity and electric conductivity measurements were carried out using a standard dc technique in the continuous-flow helium cryostat in the temperature range 1.5–170 K in magnetic fields up to 13 T. A typical six-contact sample configuration, which allows both the sample resistance and Hall voltage to be measured simultaneously, was used. Examples of the transverse resistivity ρ_{xy} dependences on the magnetic field for three samples: $Ge_{1-x}Mn_xTe$ – 960_2, $Ge_{1-x-y}Mn_xEu_yTe$ – 849_16 and $Ge_{1-x}Mn_xTe$ – 960_12 are shown in Fig. 3. The anomalous Hall effect component considerably affects the ρ_{xy} dependences for the samples $Ge_{1-x-y}Mn_xEu_yTe$ – 849_16 and $Ge_{1-x}Mn_xTe$ – 960_12. In the case of the sample with the lowest manganese content, i.e. $Ge_{1-x}Mn_xTe$ – 960_2, the anomalous Hall contribution to the transverse resistivity is practically absent.

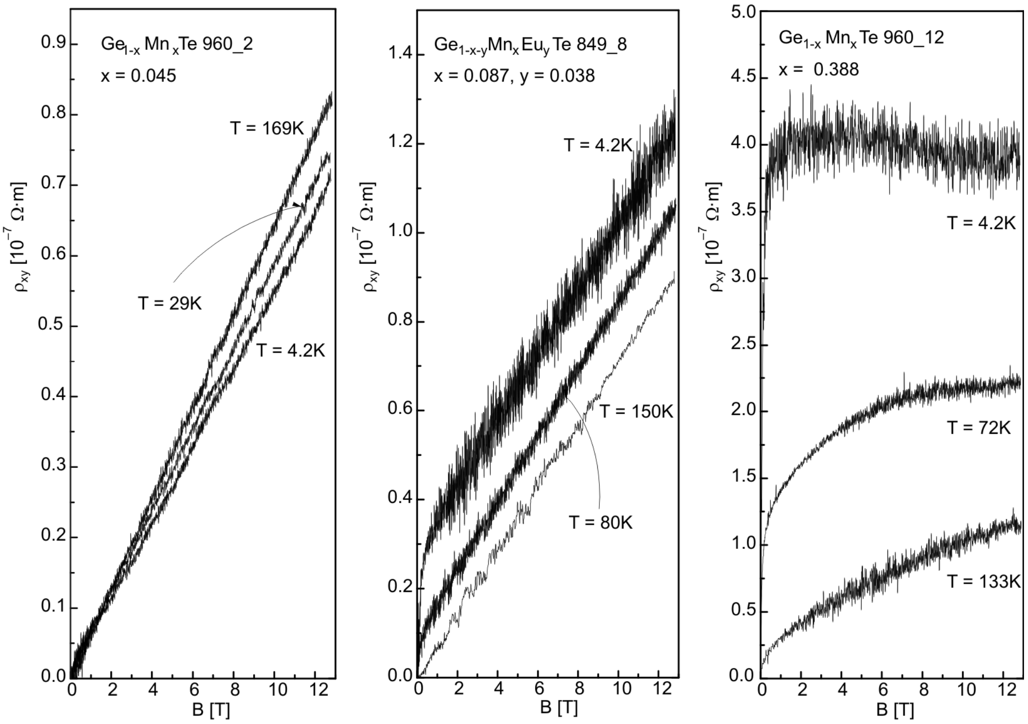

Fig. 3. Transverse resistivity of selected samples for low (left), medium (central) and high (right) manganese composition, at three chosen temperatures below and near (above) the Curie temperature

In order to extend our previous studies of the anomalous Hall effect in IV–VI mixed compounds [6, 7], the transport and magnetic measurements were performed for the same set of temperatures.

3. Results and conclusions

The ordinary and anomalous Hall coefficients were determined from the total transverse resistivity and magnetization data by the least square root fit to the equation $\rho_{xy} = R_0B + \mu_0R_SM$ [8], where B is the magnetic field, R_0 and R_S are the normal and anomalous Hall coefficients, respectively, and μ_0 is the permeability constant. In the following, we assumed that the carrier concentration in the investigated samples does not depend on the temperature in the investigated temperatures range. The strong p-type metallic conductivity justifies this assumption. We applied the two-step procedure: (i) in the first step, both Hall coefficients: R_0 and R_S were evaluated as the result of the fitting process for individual temperatures, (ii) in the second step, we fixed averaged R_0 values and repeated the fitting with R_S as the only fitting parameter. It may be noted that the quality of the fit is very good and the theoretical curves ideally overlap the experimental data. As a result of the fitting procedure, we obtained the anomalous Hall coefficient value separately for each of the temperatures. The results are presented in Fig. 4. It may be noted that except for one sample, i.e. $Ge_{1-x-y}Mn_xYb_yTe$, within the experimental errors, the anomalous Hall coefficients are temperature independent. The source of the different behaviour observed in $Ge_{1-x-y}Mn_xYb_yTe$ sample is not known. The likely reason may be the presence of the inclusions of the second crystallographic or chemical phase. It may lead to a different temperature behaviour of the anomalous Hall coefficient in that sample.

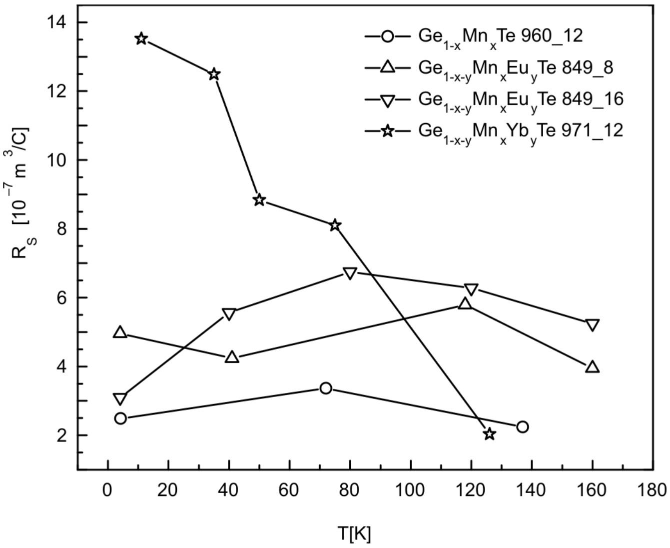

Fig. 4. Anomalous Hall coefficient R_S vs. temperature for $Ge_{1-x-y}Mn_x(Eu,Yb)_yTe$ samples

Generally, conclusions of the present work are similar to the ones derived from our previous studies on SnTe-based ferromagnetic mixed crystals [7]. It was found that, within the experimental error, the anomalous Hall coefficient R_S does not depend on temperature. However, we noted one sample in which the anomalous Hall coefficient shows an untypical behaviour. The origin of this anomaly is not yet understood. The anomalous Hall coefficient is sensitive to the carrier concentration, the increase of carrier concentration leads to a decrease of R_S value. It should also be noted that the absolute values of the anomalous Hall coefficient are of the same order in the both investigated systems, SnTe-based as well as GeTe-based.

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