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The Role of Frustration in Magnetism of $Ge_{1-x}Cr_xTe$ Semimagnetic Semiconductor

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We present preliminary studies of magnetic properties of $Ge_{1-x}Cr_xTe$ semimagnetic semiconductors with low chromium content $x < 0.026$. The static and dynamic magnetometry techniques were employed for the current investigations. The obtained results showed large bifurcations between zero-field cooled and field cooled magnetization curves at temperatures lower than 50 K. The dynamic susceptibility measurements proved via frequency shifting of the peaks that the observed magnetic order at low temperatures was the spin-glass-like state caused by magnetic frustration of the system.

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1. Introduction

In the last few years there has been generated a considerable interest in the magnetic properties of IV–VI semimagnetic semiconductors (SMSs) such as GeTe containing transition metals. Chen et al. [1], Fukuma et al. [2], and Knoff et al. [3] investigated thin films of $Ge_{1-x}Mn_xTe$ and reported their ferromagnetic properties. $Ge_{1-x}Mn_xTe$ is a promising candidate for being the first SMSs capable of being utilized for magnetic recording at room temperature due to large Curie temperatures as high as 200 K reported in this material [4, 5]. Chromium alloying is a promising step towards room temperature ferromagnetism in GeTe based semimagnetic semiconductors due to ferromagnetism observed recently in epitaxial $Ge_{1-x}Cr_xTe$ thin layers [6–8]. So far there is no literature evidence for chromium alloying of bulk GeTe crystals. However, the addition of chromium might increase the temperatures of ordering in this class of compounds and is a step towards understanding the magnetic properties of these complex alloys.

Our recent studies of bulk $Ge_{1-x-y}Sn_xMn_yTe$ mixed crystals indicated a paramagnet-spin-glass transition [9, 10]. In the present paper we present preliminary work devoted to bulk $Ge_{1-x}Cr_xTe$ crystals with chromium content x values of 0.016 and 0.025. The present investigations allowed us to determine the possibilities of tuning the magnetic properties of chromium alloyed GeTe SMSs.

2. Sample preparation and basic characterization

The studied $Ge_{1-x}Cr_xTe$ crystals were prepared using the modified Bridgman method. The modifications of the growth procedure were similar to the ones applied for the growth of alumina crystals (see Ref. [11]) and consisting of the presence of an additional radial temperature gradient in the growth furnace. The modifications changed the crystallization front of about 15 deg and decreased the number of the crystal blocks in the ingots from a few down to no more than two.

The chemical composition of the crystal blocks was determined using the energy dispersive X-ray fluorescence method. The as-grown blocks were cut into slices of about 1 mm thick perpendicular to the growth direction. We have chosen two samples for the present studies in which the amount of Cr ions was $x = 0.016$ or $x = 0.025$.

The crystallographic quality of the studied $Ge_{1-x}Cr_xTe$ samples was checked using the powder X-ray diffraction (XRD) technique. Analysis of the X-ray diffractograms showed that all the studied crystals were single phased. No phase separations, especially those containing chromium were detected. The samples had rhombohedrally distorted NaCl structure with

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lattice parameters similar to the ones for pure GeTe i.e. the lattice constant a was around 5.98 Å and the angle of distortion α was close to 88.3 deg [12].

The basic electronic properties of $Ge_{1-x}Cr_xTe$ crystals were studied by means of magnetotransport investigations including resistivity and Hall effect measurements. The obtained results were typical for narrow gap GeTe based semiconductors i.e. low resistivity $\rho_{xx} \approx 10^{-4}$ Ω cm, high p-type conductivity with high carrier concentrations $n \approx (1 \div 3) \times 10^{20}$ cm^{-3}, and low carrier mobilities $\mu \leq 200$ cm^2/(V s).

3. Magnetic investigations

The magnetic properties of $Ge_{1-x}Cr_xTe$ crystals were studied by means of static and dynamic magnetometry methods. The static magnetization measurements done at constant magnetic fields were performed using Quantum Design MPMS XL7 Squid magnetometer. Dynamic susceptibility measurements in the presence of alternating magnetic field were performed using LakeShore 7229 susceptometer system.

The magnetization M as a function of temperature was investigated using four different magnetic field H changing in the range 20–200 Oe. The results of the $M(T)$ measurements for selected $Ge_{0.984}Cr_{0.016}Te$ crystal are gathered in Fig. 1. As we can observe in Fig. 1 the magnetization in the presence of an external magnetic field rises rapidly as temperature is lowered below 50 K.

The behavior of the $M(H)$ curves was almost linear for all measured temperatures in the studied samples. The magnetization $M(H)$ curves obtained for the studied samples were used for calculating the zero-field cooled (ZFC) and field cooled (FC) temperature dependences of

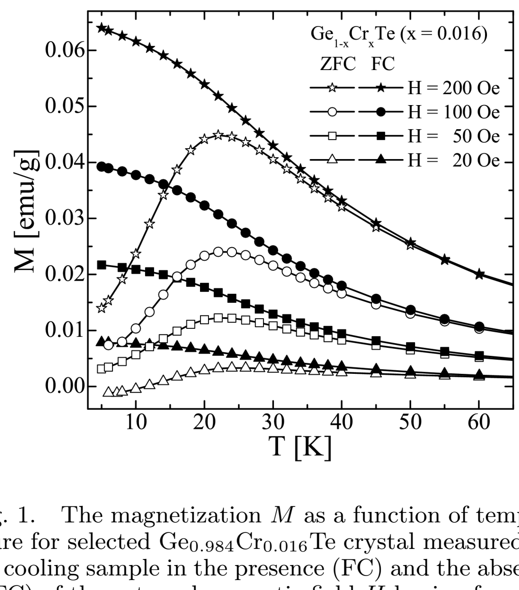

Fig. 1. The magnetization M as a function of temperature for selected $Ge_{0.984}Cr_{0.016}Te$ crystal measured after cooling sample in the presence (FC) and the absence (ZFC) of the external magnetic field H having four different amplitudes.

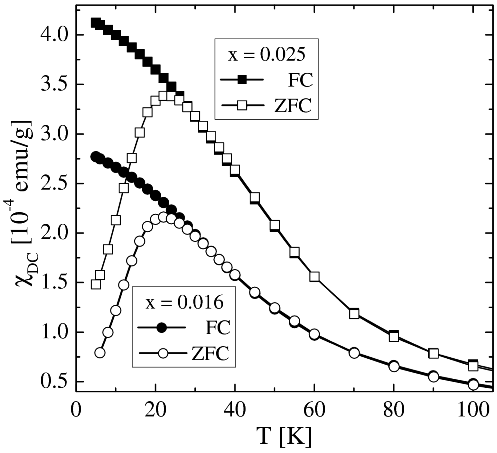

Fig. 2. The zero-field cooled and field cooled static magnetic susceptibility χ_{DC} as a function of temperature obtained for two studied $Ge_{1-x}Cr_xTe$ crystals with different chemical composition x (see labels).

the static magnetic susceptibility χ_{DC}. The results of the DC magnetic susceptibility calculations are presented in Fig. 2. As we can observe in Fig. 2 the $\chi_{DC}(T)$ curves obtained for both studied $Ge_{1-x}Cr_xTe$ crystals have similar character. The presence of broad, symmetrical cusps in ZFC $\chi_{DC}(T)$ curves as well as significant decrease of magnetic susceptibility with lowering the temperature below the cusp is a clear signature that we do not observe ferromagnetic state at low temperatures in the case of the studied crystals. We can also clearly observe large bifurcations between the observed ZFC and FC $\chi_{DC}(T)$ curves at $T < 50$ K. The observed character of the susceptibility is a typical feature of appearance of a transition to the magnetically frustrated state e.g. spin-glass or superparamagnetic ones. However, without making additional measurements it is hard to determine convincingly the type of the observed magnetic state.

The inverse of the magnetic susceptibility $(\chi_{DC})^{-1}(T)$ measured at temperatures much larger than the observed magnetic ordering ($T > 200$ K) did not vary linearly with temperature. That is, the modified Curie–Weiss law was not fulfilled in the case of the studied crystals. The susceptibility had larger values in the paramagnetic regime than it should have, assuming known chromium content. A small constant contribution from ferromagnetic CrTe clusters similar to the case of $Pb_{1-x}Cr_xTe$ in Ref. [13] was possibly observed in the studied $Ge_{1-x}Cr_xTe$ samples. However, it should be noted that the observed clusters were not detected by means of XRD.

Dynamic susceptibility measurements are an efficient tool in determining the types of magnetic states of the solid. We performed measurements of dynamic magnetic susceptibility as a function of temperature while applying an alternating magnetic field having a small amplitude $H_{AC} \leq 10$ Oe and variable frequency f changing between 7 and 9980 Hz. The results of the ac susceptibility measurements are shown in Fig. 3. As we can observe in Fig. 3 the maxima of the cusps in the ac magnetic susceptibility i.e. the freezing temperature $T_{\rm F}$, shifted towards higher temperatures with increasing frequency of the alternating magnetic field $H_{\rm AC}$. Such behavior in connection with bifurcations of static magnetization presented in Fig. 2 is a clear signature that a magnetically frustrated state is observed in the studied system. The frequency shifting of the real part of dynamic magnetic susceptibility $\chi_{\rm AC}$ can be used for determining the type of the observed magnetic state via calculation of factor proposed by Mydosh [14]:

$$R_{\rm M} = \frac{\Delta T_{\rm F}}{T_{\rm F}\log(\Delta f)}\,. \qquad (1)$$

The values of $R_{\rm M}$ factor obtained for two $Ge_{1-x}Cr_xTe$ crystals were around 0.05. The values reported for canonical spin-glass such as AuFe [14] were close to 0.01 while for a superparamagnet $R_{\rm M}$ should be higher than 0.1. Thus we find that our system is a mixed state between spin-glass and ferromagnet, which causes $R_{\rm M}$ to be higher. Our conclusions are based on high temperature results of the $\chi_{\rm DC}(T)$ measurements.

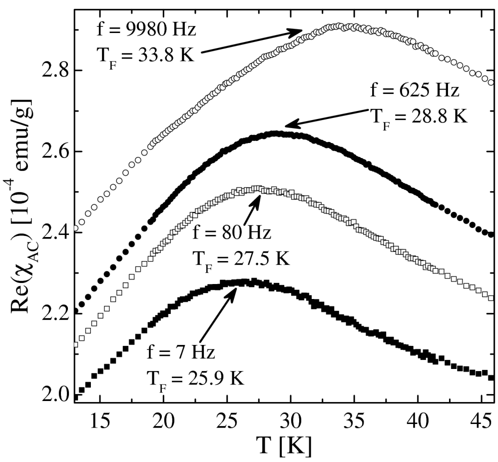

Fig. 3. The real part of the dynamic magnetic susceptibility $\chi_{\rm AC}$ as a function of temperature measured using different alternating magnetic field frequencies for selected $Ge_{0.984}Cr_{0.016}Te$ crystal.

The spin-glass transition temperature $T_{\rm SG}$ can be estimated from extrapolation of $T_{\rm F}(f)$ dependence towards $f \to 0$. The obtained $T_{\rm SG}$ values were equal to 24.2 K and 31.8 K for samples with $x = 0.016$ and 0.025, respectively. The $T_{\rm SG}$ was an increasing function of the Cr content in the samples as expected for a diluted magnetic system.

4. Summary

Preliminary investigations of magnetic properties of $Ge_{1-x}Cr_xTe$ crystals were presented. The magnetic order was observed by means of both static and dynamic susceptibility measurements in both crystals. The analysis of the experimental data proved that the spin-glass-like state with transition temperatures $T_{\rm SG}$ changing between 24.2 K and 31.8 K for crystals with $x = 0.016$ and 0.025, respectively, was observed.

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