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Colossal linear magnetoresistance in a $CdGeAs_2$:MnAs micro-composite ferromagnet

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ABSTRACT

The magnetic properties of a $Cd_{1-x}Mn_xGeAs_2$:MnAs hybrid micro-composite have the signature of ferromagnetic order at room temperature. Magnetotransport reveals the presence of a large linear positive magnetoresistance, with maximum values of about 550% ($B = 22$ T) for $x = 0.028$. This is interpreted in terms of an effective medium approximation with the value and the shape of the magnetoresistance depending on the structural and electronic properties of the semiconductor rather than on its magnetic properties. The absence of an anomalous Hall effect leads to the conclusion that ferromagnetic ordering is not the only factor necessary for the occurrence of asymmetric scattering in granular ferromagnetic materials.

1. Introduction

Since the first observation of carrier induced ferromagnetism in $Pb_{1-x-y}Sn_xMn_yTe$ at liquid helium temperatures [1], the progress in the field of ferromagnetic semiconductors (FMSs) has seen the Curie temperature (T_C) in $Ga_{1-x}Mn_xAs$ (the most intensively studied FMS at present) increase up to the current value of 185 K [2]. Due to fundamental difficulties in obtaining room temperature ferromagnetism in diluted FMSs, hybrid semiconductor–ferromagnetic metal systems have recently been intensively studied (see e.g. Refs. [3–6]). The magnetic and magnetotransport effects arising from the presence of the ferromagnetic grains in the host semiconductor matrix are interesting both from a physical and an application point of view.

The II–IV–V_2 group of chalcopyrite ferromagnetic semiconductors is a potential candidate for semiconductor spintronics [7,8]. Moreover, from an application point of view, this class of materials possesses a number of important optical properties such as large nonlinear optical coefficients and a suitable birefringence [9].

2. Sample characterization

In the following, we present structural, magnetic, and magnetotransport studies performed on $Cd_{1-x}Mn_xGeAs_2$ crystals with average chemical composition, determined using energy dispersive X-ray fluorescence (EDXRF), in the range $0.021 < x < 0.051$. The bulk $Cd_{1-x}Mn_xGeAs_2$ crystals were grown using a direct fusion method. The details of the growth procedure are presented in Ref. [10].

The crystallographic quality of the studied crystals was determined using a powder X-ray diffraction method (XRD) using a high resolution X'Pert PRO MPD diffractometer equipped with a strip detector and an incident-beam Johansson monochromator. The indexing procedure was applied to the obtained diffraction patterns in order to extract relevant crystallographic parameters of the crystals. The main phase of the crystals was identified to be a tetragonal chalcopyrite $CdGeAs_2$ structure with $I\bar{4}2d$ symmetry. The lattice parameters of all the crystals were close to the ones reported for pure $CdGeAs_2$, e.g., $a = 5.942(2)$ Å and $c = 11.224(2)$ Å [11], and changed monotonically (a decreased while c increased by around 0.2%) with increasing Mn content in the alloy. In addition to the main phase of the crystal, hexagonal and rhombohedral MnAs clusters are also detected.

Scanning electron microscope (coupled with X-ray fluorescence microprobe) studies revealed the presence of MnAs clusters with a diameter of about 1 μm in all the samples investigated (see Fig. 1). The chemical composition measured in different regions of the crystals shows that only a small fraction of Mn ions (around 1 at.%) are incorporated into the semiconductor host.

Additional confirmation of the presence of MnAs clusters in $Cd_{1-x}Mn_xGeAs_2$ crystals is provided by Nuclear Magnetic

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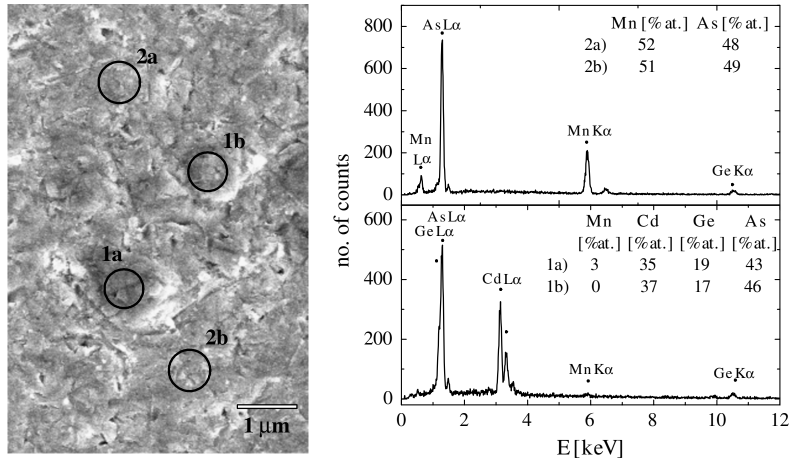

Fig. 1. Results of (a) SEM and (b) EDS measurements performed on the $Cd_{1-x}Mn_xGeAs_2$ crystal with 2.8 at.% of Mn.

Resonance (NMR) experiments. The zero-field spin-echo measurements of ^{55}Mn and ^{75}As isotopes were performed using an automated, coherent, and phase-sensitive spectrometer [12] at a temperature of $T \approx 4.3$ K. The resulting NMR spectra contained broad peaks near 210 and 240 MHz, for all the crystals investigated. NMR peaks were not observed in the frequency range in which peaks characteristic of Mn^{2+} ions in the ferromagnetic state should be detected. All the studied $Cd_{1-x}Mn_xGeAs_2$ crystals display NMR spectra characteristic of bulk ferromagnetic MnAs crystals in which peaks around 236.1 and 207.9 MHz should be observed [13].

The magnetic properties of $Cd_{1-x}Mn_xGeAs_2$ crystals were measured using a vibrating sample magnetometer (VSM). Fig. 2 shows the temperature dependence of the magnetization M for the studied samples. At temperatures below 325 K a ferromagnetic order is observed. The ferromagnetic nature of the magnetic ordering is confirmed by the hysteretic behavior of magnetization versus magnetic field curves with small coercive fields $H_C < 130$ Oe and a spontaneous magnetization $M_R < 0.2$ emu/g at temperatures below T_C. The magnetization of the samples saturates at magnetic field of about $B = 7$ T. The values of the saturation magnetization M_S varies with the Mn content between 3 emu/g for the sample with 2.1 at.% of Mn and 9 emu/g for the sample with 5.1 at.% of Mn. The estimated values of M_S (under the assumption that Mn ions have a 3d^5 configuration with spin $S = 5/2$) using average chemical content x are similar to the experimentally observed magnetization indicating that most of the Mn is magnetically active. The observed values of the Curie temperatures in the studied samples are in the temperature range $304 < T_C < 323$ K. It is known that for GaAs:MnAs systems the Curie temperature depends on the geometrical parameters and stoichiometry of MnAs clusters [3] and the same interpretation can be invoked here. In view of the fact that the NMR spectra showed no peaks originating from the dissolved manganese ferromagnetically coupled through the indirect long range interactions it is obvious to assume that most of the magnetization observed in VSM measurements reflects the intergrain ferromagnetic coupling of MnAs clusters.

3. Magnetotransport studies

The DC magnetotransport measurements were performed in high magnetic fields B up to 23 T over a wide range of temperatures $1.4 \leq T \leq 300$ K. All of the studied samples were n type and their basic electrical parameters are summarized in Table 1. The Hall carrier concentration n and electron mobility μ are between $1 \times 10^{18} \leq n \leq 2 \times 10^{20}$ cm^{-3} and $0.3 \leq \mu \leq 233$ cm^2/(Vs) at room temperature. Large changes of carrier concentration and mobility are correlated with deviations from crystal stoichiometry (detected using EDXRF method) inducing point defects which act as a source of charge and scattering centers.

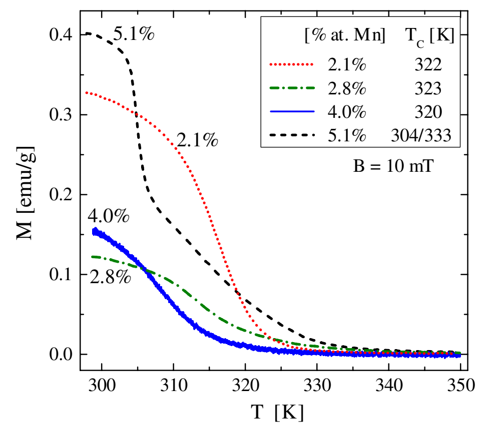

Fig. 2. The DC magnetization as a function of temperature measured for $Cd_{1-x}Mn_xGeAs_2$ samples with different chemical average composition x (shown in legend).

Table 1
Average chemical compositions x and electronic properties of $Cd_{1-x}Mn_xGeAs_2$ crystals at room temperature: resistivity ρ_{xx}, carrier concentration n and mobility μ.

x [at.%]	ρ_{xx} [10^{-2} Ω cm]	n [10^{19} cm^{-3}]	μ [cm^2/(Vs)]
2.1 ± 0.2	4.4 ± 0.1	1.13 ± 0.1	0.3 ± 0.2
2.8 ± 0.3	1.5 ± 0.1	0.102 ± 0.011	15.6 ± 0.5
4.0 ± 0.4	41.1 ± 1.5	0.179 ± 0.021	233 ± 4
5.1 ± 0.5	202.0 ± 2.0	20.1 ± 1.3	0.7 ± 0.03

Perhaps surprisingly, we find no evidence for an anomalous Hall effect (AHE) at low magnetic fields. The ρ_{xy} versus B curves are to a good approximation nearly linear up to high magnetic fields $B \approx 23$ T (see Fig. 3). The AHE is widely observed in many granular ferromagnetic systems [12,13]. The results obtained in our study indicate that despite the presence of ferromagnetism in the material an AHE does not occur. It must be stressed that in granular ferromagnetic materials the asymmetric scattering leading to the AHE cannot be clearly linked with ferromagnetic ordering of the material.

Most of the studied samples showed a positive magnetoresistance $\Delta\rho/\rho_o = (\rho(B) - \rho(B = 0))/\rho(B = 0)$. The only exception was the sample with 4.0 at.% of Mn. The highest value of magnetoresistance was observed in the sample with 2.8 at.% of Mn and

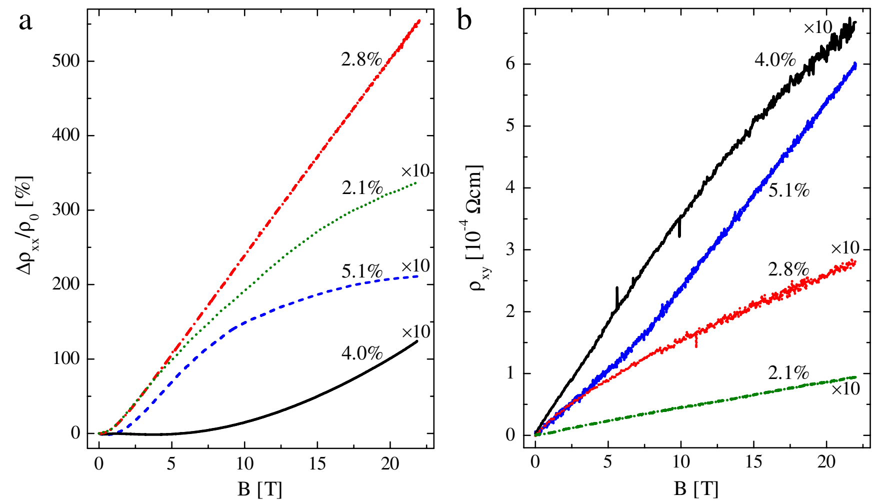

Fig. 3. The magnetic field dependence of (a) ρ_{xx} and (b) ρ_{xy} obtained at $T \approx 1.5$K for all the studied $Cd_{1-x}Mn_xGeAs_2$ samples with different average chemical composition x.

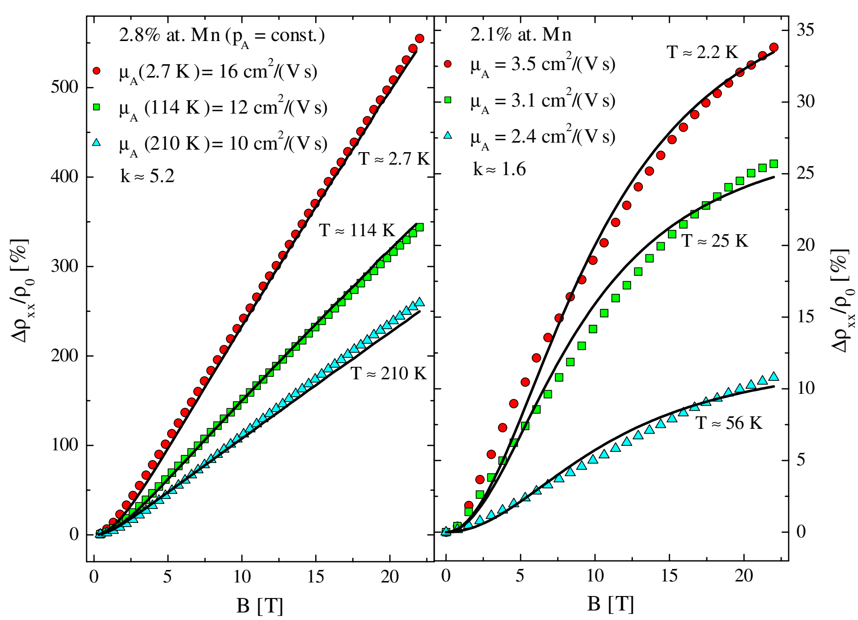

Fig. 4. The normalized resistivity ρ_{xx} as a function of a magnetic field (experimental data — symbols, theoretical curves — lines) for selected $Cd_{1-x}Mn_xGeAs_2$ samples.

reached about 550% at $T = 1.5$ K. An analysis of the results indicate that there is no correlation between amplitude of the magnetoresistance $\Delta\rho/\rho_o$ and the average composition x but there exists a relation between the Hall mobility of the sample and $\Delta\rho/\rho_o$. The highest value of positive $\Delta\rho/\rho_o$ was observed for the sample with $\mu \approx 16$ cm^2/(Vs). For the sample with the highest mobility $\mu \approx 230$ cm^2/(Vs) only a small negative magnetoresistance was observed, possibly caused by a weak localization effect. The high carrier mobility may enhance quantum effects while masking the classical effects [14]. The large positive magnetoresistance was observed recently in a number of inhomogeneous granular systems *e.g.* [15–17]. The effect reported in the present work has one of the highest observed amplitudes and persists with a large value $\Delta\rho/\rho_o \approx 250\%$ even at $T \approx 210$ K (see Fig. 4).

The magnetoresistance of a two phase solid with different electronic properties can be described theoretically using the effective medium approximation (EMA) model [18,19]. This model allows the calculation of the effective conductivity tensor components $\sigma_{A,B}$ of two crystal phases A and B having concentrations $p_A = 1 - p_B$ and mobility ratio $k = \mu_A/\mu_B$. The magnetic field dependence of resistivity may be calculated by solving the self-consistent equation:

$$\sum_{i=A,B} p_i \delta\sigma_i (I - \Gamma\delta\sigma_i)^{-1} = 0, \tag{1}$$

where Γ is a depolarization tensor describing the mean shape of the cluster. The EMA model was used in order to fit a theoretical magnetoresistance curve to the experimental one, with μ_B taken as a fitting parameter. The Hall carrier mobility of the semiconductor matrix μ_A was taken as fixed but temperature dependent parameter having values equal to the obtained ones from the Hall effect measurements. Since the $\rho_{xy}(B)$ curves were nearly linear for all the studied temperatures (see Fig. 3(b)) up to magnetic field $B \approx 23$ T we can assume that the carrier transport in the studied material is dominated by the scattering in semiconductor host. In this case the value of the Hall carrier mobility μ reflects the carrier mobility in the semiconductor matrix. The results of the theoretical fit are presented in Fig. 4. Clearly the EMA model well reproduces the shape of the observed experimental results and also provides a good quantitative agreement for the size of the magnetoresistance. It should be noted that an increase of the ratio *k* leads to a higher value of the magnetoresistance and to a more pronounced linearity.

4. Summary

In conclusion, a colossal positive magnetoresistance (550%) has been observed in the $CdGeAs_2$:MnAs hybrid semiconductor–metal granular ferromagnet and can be interpreted in terms of an effective medium approximation. The observed phenomenon is due to the structural properties of the samples and depends on the ratio between the mobility of the individual phases present in the crystal. There is no apparent correlation between the magnetic properties and the size of the magnetoresistance. The absence of an AHE in the studied material is a clear signature that ferromagnetic order is not the only element required to induce asymmetric scattering in the material. The preliminary results presented in this work offer hope that the optimally processed crystals (e.g. via post-growth heat treatment) can effectively combine room temperature ferromagnetism with colossal magnetoresistive effect and also have interesting optical properties.

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