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MAGNETIC SPECIFIC HEAT STUDY OF THE COMPETITION BETWEEN FERRO- AND ANTIFERROMAGNETIC SPIN-SPIN EXCHANGE INTERACTIONS IN PbSnMnTe*

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Magnetic contribution to the specific heat, magnetic susceptibility and Hall effect are experimentally studied in $Pb_{1-x-y}Sn_yMn_xTe$ semimagnetic semiconductors with $y = 0.72$ and $x = 0.08$ and with different carrier concentrations $10^{20} \leq p \leq 10^{21}$ cm^{-3}. The ferromagnetism observed in crystals with $p > 3 \times 10^{20}$ cm^{-3} breaks down with a decreasing concentration of carriers due to an increasing competition between Ruderman-Kittel-Kasuya-Yoshida and superexchange interactions.

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$Pb_{1-x-y}Sn_yMn_xTe$ semimagnetic semiconductors form a model diluted magnetic system with metallic electrical properties. Experimental and theoretical studies of magnetic properties of these crystals revealed a decisive role of concentration of carriers p and an important role of a two-band structure of the valence band [1-3]. The results are summarized in the form of (x, p, T) magnetic phase diagram containing both ferromagnetic (FM) and spin-glass (SG) regions depending on the ratio x/p, where x is the concentration of Mn ions [2, 3]. The spin-spin interaction mechanism responsible for these magnetic properties is the Ruderman-Kittel-Kasuya-Yoshida (RKKY) indirect exchange interaction via spin polarization of carriers. The strength of the RKKY interaction decreases with decreasing carrier concentration. In the crystals with $p \leq 10^{20}$ cm^{-3} the antiferromagnetic superexchange (SE) via anions is expected to dominate the spin-spin interactions as is observed in II-VI semimagnetic semiconductors. In IV-VI semimagnetic semiconductors one can control the concentration of carriers by annealing in an appropriate atmosphere. That offers the possibility to change the strength of the RKKY interaction leaving the SE mechanism practically unchanged. The aim of our work is to use this possibility to experimentally study the evolution of the magnetic properties of PbSnMnTe as a function of the relative contribution of ferro- and antiferromagnetic interactions.

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We studied the temperature dependence of magnetic contribution to the specific heat, magnetic susceptibility and transport properties of bulk crystals of $Pb_{1-x-y}Sn_yMn_xTe$ with $y = 0.72$ and $x = 0.08$ and with a carrier concentration in the range $10^{20} \leq p \leq 10^{21}$ cm^{-3}. The specific heat was measured in the temperature range 0.4 K$\leq T \leq$ 20 K by standard heat-pulse method. The measurements of the ac magnetic susceptibility were performed in the temperature range 1.5 K $\leq T \leq$ 70 K by mutual inductance bridge. The Hall effect was measured in the temperature range 4.2 K $\leq T \leq$ 300 K by 4-probe dc technique. In view of the quite strong anomalous Hall effect observed in our samples at helium temperatures, we characterize all our crystals by the carrier concentration at T = 77 K [4]. Our crystals were examined by the microprobe analysis and the X-ray Debye method and found to be single-phase and homogeneous.

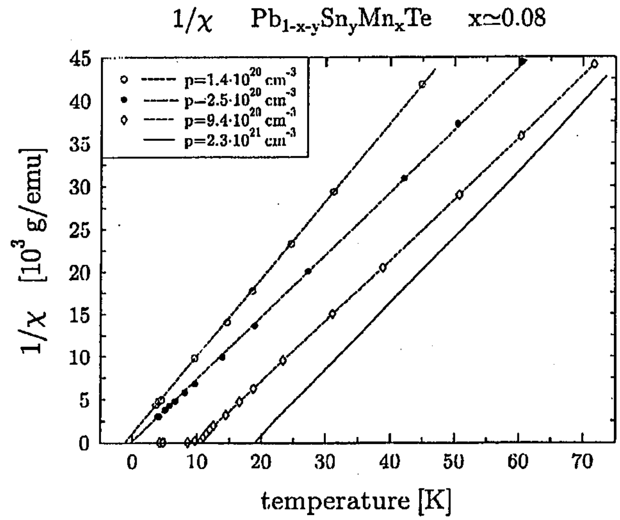

Fig. 1. The temperature dependence of the inverse magnetic susceptibility of the crystals of $Pb_{1-x-y}Sn_yMn_xTe$ with $x = 0.08$ and $y = 0.72$ and different carrier concentrations. The lines with data points present results of our studies whereas the solid line shows the behavior of the sample studied in Ref. [5].

The results of the measurements of the magnetic susceptibility are presented in Fig. 1. We found, as expected, that the samples with a carrier concentration $p > 3 \times 10^{20}$ cm^{-3} reveal the ferromagnetic properties as evidenced by the critical temperature dependence of the magnetic susceptibility and by the observation of the well-defined magnetic contribution to the specific heat with a maximum slightly below the Curie temperature derived from the analysis of the temperature dependence of the magnetic susceptibility. The samples with a lower carrier concentration, which are of our primary concern, remain paramagnetic in the whole temperature region covered by our susceptometer, i.e. down to $T = 1.5$ K.

The temperature dependence of the inverse magnetic susceptibility of the sample with $p = 2.5 \times 10^{20}$ cm^{-3} follows the Curie–Weiss law with the paramagnetic Curie temperature $\Theta \approx 0$. Θ is given by the well-known relation $k_B\Theta = (2/3) \times xS(S+1)\sum z_i I(R_i)$, where $I(R_i)$ is the exchange interaction between the two spins at the distance R_i and z_i is the number of lattice positions in the i-th crystallographic shell. The $\Theta = 0$ indicates an (apparent) lack of interactions between the spins which in our case is simply the manifestation of the mutual compensation of the contributions from the RKKY and the SE mechanisms. The sample with $p = 1.4 \times 10^{20}$ cm^{-3} shows $\Theta = -1.1$ K, i.e. of antiferromagnetic sign.

To verify whether these samples undergo a magnetic phase transition at lower temperatures we have performed the specific heat measurements at subkelvin temperatures. The results are presented in Fig. 2a together with the data for ferromagnetic sample studied in Ref. [5] presented in Fig. 2b. The magnetic contribution to the total specific heat was determined as the difference between the experimentally measured total (C_t) specific heat and the crystal lattice contribution (C_{lat}) determined based on the $C_t \sim T^3$ dependence observed at higher temperatures. Both samples show no indication for a subkelvin ferromagnetic tran-

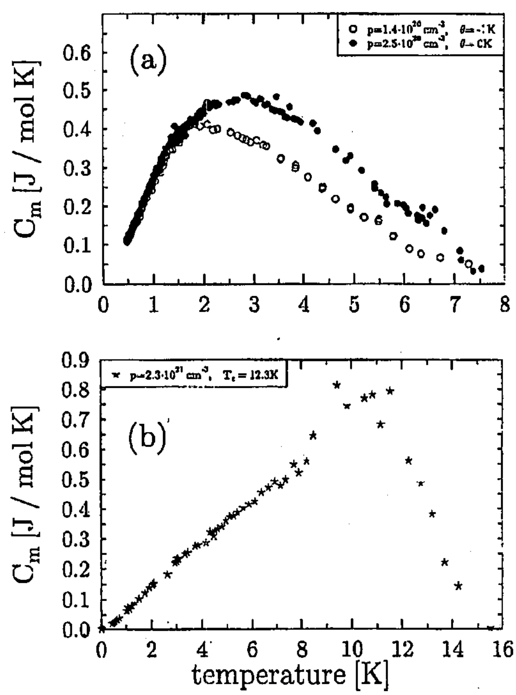

Fig. 2. The temperature dependence of the magnetic contribution to the specific heat of the samples $Pb_{0.2}Sn_{0.72}Mn_{0.08}Te$ with different carrier concentrations (a); (b) shows the magnetic contribution to the specific heat of the ferromagnetic sample studied in Ref. [5].

sition. The observed temperature dependence of the specific heat cannot also be explained within the frames of the cluster models predicting a more flat temperature dependence. The broad contribution to the magnetic specific heat with the linear low temperature behavior indicates rather a formation of a disordered magnetic phase of spin-glass-type. This result can be easily understood as both crystals reveal two essential features of a spin-glass system, i.e., the disorder and the frustration leading to the competition between the exchange couplings of different strength and different sign.

The physical situation in these samples is quite different. The sample with $p = 1.4 \times 10^{20}$ cm^{-3} seems to have the concentration of carriers low enough to neglect the RKKY mechanism. The important factor here is the location of the Fermi level above the band of heavy holes what strongly reduces the RKKY interaction [1]. This sample is a magnetic analog of, e.g. $Cd_{1-x}Mn_xTe$ and represents the system with dominant antiferromagnetic interaction coupling the spins allocated over the sites of fcc magnetic sublattice with the inherent topological frustration leading to the spin-glass-like behavior. In the case of the sample with $p = 2.5 \times 10^{20}$ cm^{-3} ($\Theta \approx 0$) we expect that more important will be the effect of the competition between the interactions of different signs. Apart from the shift of the maximum on the temperature dependence of the magnetic specific heat, there seems to be no qualitative difference between the two above discussed non-ferromagnetic samples studied by us. It indicates that this is rather the similar disordered magnetic structure, not the details of the exchange interactions, what determines the temperature dependence of the magnetic contribution to the specific heat. The definite conclusions about a spin-glass formation in low carrier concentration samples and not, e.g. a complicated super-paramagnetic behavior, can only be reached from the measurements of the magnetic susceptibility and the magnetization at $T < 1.5$ K. Such experimental data have not been available yet.

In conclusion, our results give the first experimental evidence for two mechanisms creating a spin-glass-like phase in PbSnMnTe being of different origin than the RKKY spin-glass (observed earlier in samples with a very high carrier concentration and relatively low Mn content [3]). The first one results from the competition between the spin–spin interactions of different sign (the case of $\Theta \approx 0$) and the second one from the topological frustration effects present in the systems with only antiferromagnetic interactions (the case of negative Θ).

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