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Ferromagnetic Transition in $\mathbf{Ge_{1-x}Mn_xTe}$ Layers

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Ferromagnetic transition temperature in thin layers of diluted magnetic (semimagnetic) semiconductor $Ge_{1-x}Mn_xTe$ was studied experimentally by SQUID magnetometry method and analyzed theoretically for a model Ising-type diluted magnetic system with Ruderman–Kittel–Kasuya–Yosida indirect exchange interaction. The key features of the experimentally observed dependence of the Curie temperature on Mn content ($x \leq 0.12$) and conducting hole concentration $p = (1\text{–}10) \times 10^{21}$ cm^{-3} were reproduced theoretically for realistic valence band and crystal lattice parameters of p-$Ge_{1-x}Mn_xTe$ taking into account short carrier mean free path encountered in this material and Ruderman–Kittel–Kasuya–Yosida mechanism with both delta-like and diffused character of spatial dependence of the exchange coupling between magnetic ions and free carriers.

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1. Introduction

In IV–VI diluted magnetic (semimagnetic) semiconductors with magnetic ions of Mn ferromagnetic transition is induced by a very high concentration of conducting holes and is usually related to the well-known Ruderman–Kittel–Kasuya–Yosida (RKKY) indirect exchange interaction mechanism. In diluted magnetic systems with the RKKY exchange interaction, increasing the carrier concentration changes not only the critical temperature of magnetic transition T_{C} but also switches the character of magnetic ordering. The analysis of these effects in $Sn_{1-x}Mn_xTe$ and $Pb_{1-x-y}Sn_yMn_xTe$ bulk crystals showed that ferromagnetic, spin glass as well as re-entrant spin glass phases are experimentally found in the x–p magnetic phase diagram for the examined Mn ions content $x \leq 0.12$ and conducting hole concentration $p \approx 10^{20}$–10^{21} cm^{-3} [1, 2]. $Ge_{1-x}Mn_xTe$ is a distinct IV–VI semimagnetic semiconductor that exhibits the highest (in this materials family) ferromagnetic transition with the Curie temperature up to $T_{\mathrm{C}} \approx 150$ K as found in early studies carried out on bulk polycrystals [3] and up to $T_{\mathrm{C}} \approx 190$ K as observed in recent studies of epitaxial layers [4, 5]. This is primarily related to relatively high p–d exchange coupling $J_{pd} = 0.4$–0.8 eV reported in GeMnTe as compared to $J_{pd} = 0.05$–0.1 eV found in SnMnTe or PbSnMnTe and Fermi energy $E_{\mathrm{F}} = 0.2$–0.3 eV typically encountered in these materials [1–5].

In this work, we use superconducting (SQUID) magnetometry method to experimentally study the dependence of the ferromagnetic Curie temperature on conducting hole concentration and magnetic ions content $T_{\mathrm{C}}(x, p)$ and compare experimental findings with theoretical calculations of the Curie temperature performed for a model diluted magnetic system with the RKKY exchange interaction using mean field Ising model with realistic valence band structure parameters of GeMnTe.

2. Experimental

The layers of $Ge_{1-x}Mn_xTe$ were grown by molecular beam epitaxy on insulating $BaF_2(111)$ substrates. To avoid possible chemical and electrical homogeneity problems we limited ourselves to the layers with Mn composition in the range $x \leq 0.12$ grown under varying Te stoichiometry conditions [6]. All layers are about 1 μm thick. Our aim was to prepare two sets of layers: (1) with varying Mn content but similar conducting hole concentration and (2) with approximately constant Mn content but varying hole concentration. Although the control of Mn concentration is relatively easy in MBE grown GeMnTe layers, their electrical properties are very sensitive to even small changes in molecular fluxes of GeTe, Mn and Te. X-ray diffraction, secondary ion mass spectroscopy, energy dispersive X-ray fluorescence, and atomic force microscopy structural and chemical characterization of the layers confirmed their good monocrystallinity with rhombohedral structure of [111]-crystal axis along the layer growth direction [6, 7]. Hall effect measurements carried out at temperature $T = 77$ K revealed p-type conductivity with a very high conducting hole concentration varying in the range $p = (1\text{–}10) \times 10^{21}$ cm^{-3}.

Magnetization measurements were performed in the temperature range $T = 5\text{–}120$ K and magnetic fields up to 1.5 kOe using superconducting (SQUID) magnetometer with the field applied in the plane of the layer along [110] direction. The typical examples of the temperature dependence of magnetization and magnetic hysteresis loops observed in the GeMnTe layers are presented in Fig. 1. The ferromagnetic transition was clearly observed in all the layers studied with the Curie temperature in the range $T_C = 9\text{–}45$ K, depending on Mn content and hole concentration. The diamagnetic contribution due to BaF_2 substrates is very small as evidenced in Fig. 1b for magnetization measurements at $T = 100\ K > T_C$.

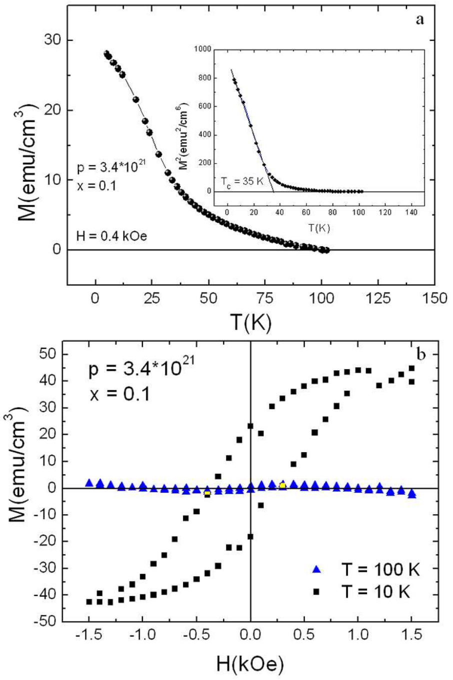

Fig. 1. (a) The temperature dependence of magnetization $M(T)$ of $Ge_{0.9}Mn_{0.1}Te$ layer. The inset illustrates the simple method of the determination of the ferromagnetic Curie temperature based on the extrapolation of the M^2 vs. T dependence. Part (b) shows magnetic hysteresis loop for the same layer measured at $T = 10$ K.

The $M(T)$ dependence typically observed in GeMnTe monocrystalline layers reveals a temperature transition region much wider that expected for materials with long ranged exchange interaction obeying the mean field description. This experimental feature is particularly strong in monocrystalline layers with low carrier concentration but appears to be of minor importance e.g. in polycrystalline materials. It strongly obscures precise experimental determination of the Curie temperature. In this work, we use a conservative T_C estimation based on the mean field extrapolation: $M^2(T) = T_C - T$ for $T < T_C$. This is consistent with the results of ferromagnetic resonance measurements on GeMnTe layers [7]. However, certain ferromagnetic features of presumably short-range character persist to much higher temperatures.

3. Theoretical model

Thorough theoretical analysis of magnetic properties of magnetic semiconductors requires, in particular: (1) identifying the dominant interspin exchange interaction mechanism, (2) establishing magnetic ground state by analyzing the magnetic energy of various spin configurations, (3) calculating magnetic transition temperature, and (4) calculating thermodynamic properties, e.g. the temperature dependence of magnetization.

In this short communication we report on the theoretical calculations of the Curie temperature of GeMnTe layers in the carrier concentration and Mn content range studied by us experimentally. We considered a model diluted magnetic system of localized Ising spins randomly occupying fcc crystal lattice positions and coupled via the long-range RKKY exchange interaction. For the RKKY interaction analysis we studied both the standard case of delta-like contact potential of the exchange coupling between the spin of local magnetic moment and the spin of conducting carriers as well as the case of, so-called, diffused contact potential. In the latter case the Gaussian probability distribution centered at the local moment site was assumed with the standard deviation parameter $\sigma = 0.7$ Å i.e. of the order of the radius of $3d$ magnetic orbital of Mn. The details of this theoretical model are discussed in Ref. [8] and [9]. The results of the calculations are presented in Fig. 2 for the case of realistic band structure parameters of GeMnTe with 4 equivalent L-point energy valleys and density of states effective mass $m^* = 2.8 m_0$. The effect of strong electronic disorder present in GeMnTe was taken into account via the RKKY interaction exponential damping factor with carrier mean free path of 1 nm. We also included the nearest neighbor Mn–Mn antiferromagnetic superexchange interactions known to operate in diluted magnetic semiconductors ($I_{AF} = -8$ K). The main parameter of the model is the p–d exchange coupling integral $J_{pd} = 0.6$ eV.

4. Discussion and conclusions

The RKKY model calculations presented in Fig. 2 reproduce the key experimental observations: a linear increase of transition temperature with Mn content $T_C(x) = 350\,x$ and nonmonotonic $T_C(p)$ dependence. Although the $T_C \propto x$ dependence is expected in mean field models with various exchange interaction mechanism, the $T_C(p)$ dependence can be related to the oscillatory change of interaction sign characteristic for the RKKY mechanism. The model semi-quantitatively accounts for the experimentally observed values of the Curie temperatures for the p–d exchange integral $J_{pd} = 0.6$ eV. As this value of J_{pd} is an almost order of magnitude larger than found in other IV–VI diluted magnetic (semimagnetic) materials the applicability of the RKKY model to GeMnTe still needs further analysis.

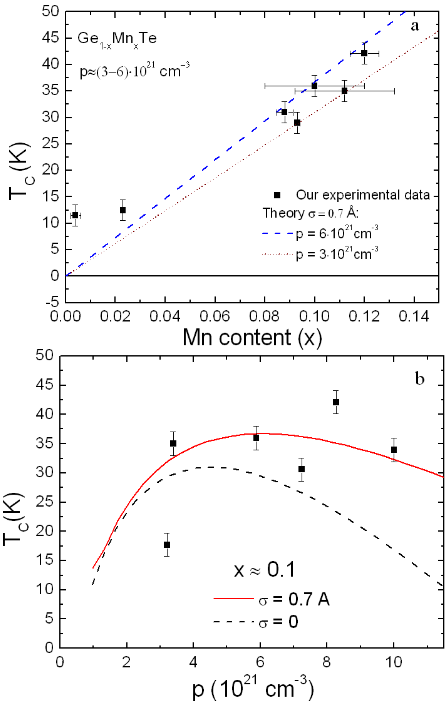

Fig. 2. The experimental and theoretical analysis of the dependence of the ferromagnetic Curie temperature of $Ge_{1-x}Mn_xTe$ layers on Mn content $x \leq 0.12$ (a) and conducting hole concentration $p = (1–10) \times 10^{20}$ cm^{-3} (b). Squares represent experimental data obtained based on the magnetization $M(T)$ analysis shown in Fig. 1a. Lines show the results of theoretical mean field calculations with various RKKY interaction models (as explained in Sect. 3).

In conclusion, employing SQUID magnetometry method we experimentally determined Mn content (x) and carrier concentration (p) dependence of the Curie temperature $T_C(x,p)$ of ferromagnetic transition observed in $Ge_{1-x}Mn_xTe$ layers ($x \leq 0.12$) with varying hole concentration (p). Theoretical analysis of the transition temperature $T_C(x,p)$ was carried out in the RKKY model for randomly diluted magnetic system of localized moments taking into account realistic band structure parameters of $Ge_{1-x}Mn_xTe$. The model correctly accounts for key experimental observations: nonmonotonic $T_C(p)$ dependence and linear increase of the Curie temperature $T_C(x)$ with increasing Mn content.

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