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CORRELATION BETWEEN MAGNETIC AND ELECTRONIC PROPERTIES OF $Sn_{1-x}Gd_xTe$*

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Magnetic susceptibility, electron paramagnetic resonance and transport properties of $Sn_{1-x}Gd_xTe$ with $0.04 < x < 0.07$ and hole concentrations in the range from 0.7×10^{20} to 16×10^{20} cm^{-3} were investigated. After annealing of the $Sn_{1-x}Gd_xTe$ samples with $x < 0.05$ in Sn vapor their hole concentration decreased from 5×10^{20} cm^{-3} to about 3×10^{20} cm^{-3} and their paramagnetic Curie temperature increased a few times. In samples with $x > 0.05$ no significant change in the magnetic properties was observed after annealing, even at lower hole concentrations. The results can be explained by assuming that an indirect exchange interaction, $4f$–$5d$-band electrons, is responsible for the coupling among Gd ions.

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1. Introduction

Magnetic properties of Bridgman-grown $Sn_{1-x}Gd_xTe$ have been investigated previously [1, 2]. The results showed a very weak antiferromagnetic exchange coupling among Gd ions, with no evidence for RKKY-type ferromagnetic interaction observed in $Sn_{1-x}Mn_xTe$ [3] and $Pb_{1-x-y}Sn_yMn_xTe$ [4]. However, in the as-grown samples the Gd content and carrier concentration were nearly inversely proportional. Here we are reporting measurements of magnetic properties of $Sn_{1-x}Gd_xTe$, for a series of values of x, with carrier concentration varying over an order of magnitude for a given Gd content.

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2. Experiment

The samples of $Sn_{1-x}Gd_xTe$ were cut from a boule grown by the Bridgman technique. The nominal x values varied from 0.05 to 0.09. The samples were annealed isothermally at about 700°C in Sn or Te atmosphere, in order to reduce or increase their carrier concentration, respectively.

The carrier concentrations and Hall mobilities were determined by standard Hall effect and conductivity measurements over a temperature range from 4.2 to 300 K, with an accuracy of about 15%. All the samples were p-type. The hole concentrations were in the range from 0.7×10^{20} to 16×10^{20} cm^{-3} and the mobilities from 4 to 300 $cm^2/(V\ s)$ at 77 K.

The magnetic susceptibility was measured by the ac mutual inductance technique in alternating magnetic fields up to 40 Oe, over a temperature range from 1.5 to 70 K.

3. Results and discussion

The susceptibility data were fitted over the temperature range from 10 to 70 K to the Curie–Weiss law. The Curie constant and paramagnetic temperature, θ, were fitting parameters. The diamagnetic susceptibility of the host lattice was taken from the susceptibility measurements of SnTe as -5×10^{-7} emu/g. The effective content of Gd ions, $\bar{x}$, i.e. the content in the cation sublattice of magnetic ions with a spin equal to 7/2, and the nearest neighbor exchange parameter J/k_B (k_B is the Boltzmann constant), were determined from the Curie constant and θ as described in Ref. [2], with estimated errors of about 20%. The effective Gd content was usually smaller than the nominal one, as in other diluted magnetic semiconductors.

In Fig. 1 we show the paramagnetic Curie temperature over the effective Gd content, $\theta/\bar{x}$, as a function of the carrier concentration for different samples. We see that for crystals with $\bar{x} < 0.05$, $\theta/\bar{x}$ at $p \approx 3 \times 10^{20}$ cm^{-3} is a few times larger than at $p > 5 \times 10^{20}$ cm^{-3}. In these samples we also observed a characteristic cusp at about 1.6 K in the temperature dependence of the magnetic susceptibility, which is an evidence of a spin glass phase. The hole concentration could not be reduced below 3×10^{20} cm^{-3} by further annealing. In samples with $\bar{x} > 0.05$ the hole concentration could be reduced below 10^{20} cm^{-3}, but no significant increase in θ with decreasing p was observed, and no cusp in the susceptibility appeared. In addition, the hole mobilities at $p \approx 3 \times 10^{20}$ cm^{-3} were almost an order of magnitude higher in samples with $\bar{x} < 0.05$ than in samples with $\bar{x} > 0.05$. The EPR measurements revealed the $g = 2.0$ resonance for all samples.

The experimental findings presented above can be explained by a model assuming that the Gd 5d energy state is degenerate with the valence band of $Sn_{1-x}Gd_xTe$. Schematic diagram of the energy levels and densities of states in samples with $\bar{x} < 0.05$ is shown in Fig. 2. The exchange coupling among Gd ions is due to an intraatomic 4f–5d exchange and interatomic 5d–5d exchange via conducting holes, and is inversely proportional to $E_{5d} - E_F$ [5]. Since θ is proportional to the sum of exchange constants over the crystal, the resonant enhancement of θ

in samples with $\bar{x} < 0.05$ at lowest hole concentration may be caused by E_F approaching E_{5d}. That confirms the proposed mechanism of exchange interaction.

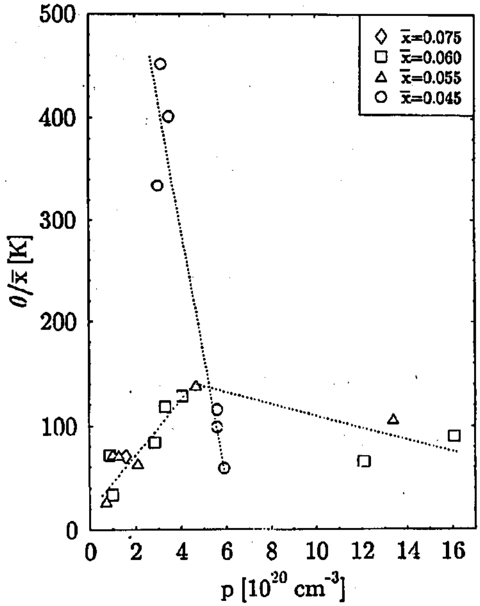

Fig. 1. $\theta/\bar{x}$ vs. hole concentration in $\mathrm{Sn}_{1-x}\mathrm{Gd}_x\mathrm{Te}$. Each $\bar{x}$ is the effective Gd content determined from susceptibility measurements and averaged over few samples, with an estimated error ±0.004. Dotted lines are a guide to the eye.

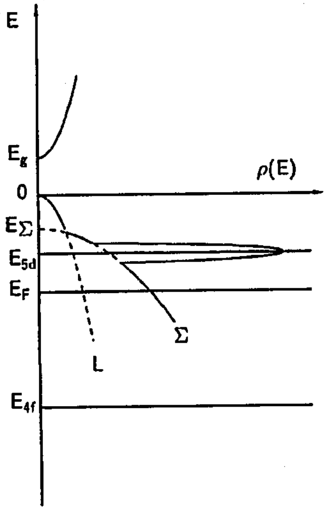

Fig. 2. Schematic diagram of energy levels and densities of states in $\mathrm{Sn}_{1-x}\mathrm{Gd}_x\mathrm{Te}$.

We believe that in samples with $\overline{x} > 0.05$ the Gd 5*d* level is degenerate only with the light hole *L*-band, the Fermi level is far below the Gd 5*d* level and practically all Gd ions are ionized Gd^{3+} ions with the outer electron configuration $4f^7$. The Fermi level may then raise relatively high in the valence band, resulting in the low hole concentration, but is still too far from the Gd 5*d* level to cause a significant increase in the exchange coupling. In samples with $\overline{x} < 0.05$ the 5*d* energy state is degenerate both with the heavy hole Σ-band and the light hole *L*-band, as shown in Fig. 2. The Fermi level may then move close to the Gd 5*d* level, causing the resonant enhancement in θ. A significant fraction of Gd ions would then be Gd^{2+} ions, with the outer electron configuration $4f^7 5d^1$. Since no significant dependence of $\overline{x}$ on hole concentration was observed, the 5*d* level is probably delocalized, and the magnetic moment of Gd in both charge states is 7/2, related only to the half-filled 4*f* shell.

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