Exchange interaction in rare-earth-doped IV-VI diluted magnetic semiconductors

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The magnetization and magnetic susceptibility of Bridgman-grown $Sn_{1-x}Gd_xTe$ samples with values of x up to 0.09 have been measured in fields up to 5.5 T over a temperature range from 2 to 300 K and in fields up to 23 T at 4.2 K. The low-field, high-temperature susceptibility data followed the Curie-Weiss relation with a small Curie temperature. The magnetic-field dependence of the magnetization was fitted to an expression containing a Brillouin function, representing isolated magnetic ions, plus a term representing pair interactions. Both the susceptibility and the high-field magnetization data indicated a weak antiferromagnetic coupling among Gd ions. These results are compared with our previously obtained exchange parameters for $Pb_{1-x}Gd_xTe$ and $Pb_{1-x}Eu_xTe$. The exchange interaction in $Sn_{1-x}Gd_xTe$ is larger than in PbTe-based, rare-earth-doped chalcogenides, in agreement with the smaller cation-anion spacing in SnTe-based compounds. A ferromagnetic or spin-glass ordering due to the possible Ruderman-Kittel-Kasuya-Yosida interaction in $Sn_{1-x}Gd_xTe$ with high hole concentration was not observed.

I. INTRODUCTION

Magnetic properties of IV-VI solid solutions containing rare-earth ions have been investigated recently, though not as widely as those of transition-metal-doped II-VI and IV-VI diluted magnetic semiconductors (DMS).[1] Mathur *et al.* measured the magnetic susceptibility of $Sn_{0.97-x}Eu_xTe$ and observed a weak antiferromagnetic coupling among Eu ions.[2] Savage and Rhyne measured the magnetic susceptibility of $Pb_{1-x}Gd_xTe$ and $Sn_{1-x}Gd_xTe$ and found a small antiferromagnetic exchange interaction in both materials.[3] Bruno *et al.* measured magnetization and magnetic susceptibility of $Pb_{1-x}Gd_xTe$ (Refs. 4 and 5) and Braunstein *et al.* have reported magnetization and susceptibility measurements on (molecular-beam epitaxy) MBE-grown thin films of $Pb_{1-x}Eu_xTe$.[6] In our previous papers we reported on the magnetic susceptibility and magnetization of $Pb_{1-x}Gd_xTe$ (Refs. 7 and 8) and $Pb_{1-x}Eu_xTe$.[9] The data have been explained by taking into account isolated rare-earth ions and pairs with contributions from larger clusters being nearly negligible for the small values of x that were investigated. The nearest-neighbor exchange interactions that were obtained were quite small, similar to those in other IV-VI DMS with the NaCl structure.

In the present paper we investigated magnetic susceptibility and magnetization of $Sn_{1-x}Gd_xTe$. Some preliminary data have been recently reported.[10] Since SnTe has a lattice constant about 2% less than PbTe, we were able to test the dependence of the exchange interaction of Gd ions on cation-anion separation in the NaCl-type lattice. Also, there is a possibility of a Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange interaction via free carriers in SnTe-based DMS with high carrier concentration. Such effects have been observed in $Sn_{1-x}Mn_xTe$ (Refs. 11–15) and $Pb_{1-x-y}Sn_yMn_xTe$.[16–19]

In the following sections we present the experimental results and analysis of low-field susceptibility and high-field magnetization measurements in $Sn_{1-x}Gd_xTe$ with $x \leq 0.09$. The exchange parameter values will be discussed and compared with those previously obtained by us for $Pb_{1-x}Gd_xTe$ and $Pb_{1-x}Eu_xTe$.

II. EXPERIMENT

Measurements of magnetization were carried out on $Sn_{1-x}Gd_xTe$ single crystals cut out from a large boule grown by the Bridgman technique. The samples were cut in the shape of Hall bars with typical dimensions of $1.5\times2\times5$ mm^3. The x values in the samples were determined by electron microprobe measurements, with an uncertainty of about 20%, which includes the variation throughout the sample. All samples were p type with carrier concentrations strongly dependent on the Gd content, changing from 1.5×10^{20} cm^{-3} in samples with the

TABLE I. Susceptibility fitting parameters and carrier concentrations for $Sn_{1-x}Gd_xTe$.

Sample	x_v	$\bar{x}$	T (K) Fit range	Θ (K)	χ_0 (emu/g)	J/k_B (K)	p (10^{20} cm^{-3})[a]
A	0.09	0.074	20–300	5.22	-5×10^{-7}	−0.56	1.58
B	0.08	0.057	20–300	6.04	-5×10^{-7}	−0.84	4.07
C	0.06	0.043	20–300	4.19	-5×10^{-7}	−0.77	5.60
D	0.05	0.039	20–300	3.46	-5×10^{-7}	−0.69	5.60
E	0.01	0.011	10–125	0.51	-1×10^{-6}	−0.36	10.0
F	0.006	0.005	10–125	0.63	-7.7×10^{-7}	−0.93	12.6

[a]Values at 77 K.

highest amount of Gd to 1.3×10^{21} cm^{-3} in samples with the smallest amount. The change in concentration is due to the fact that gadolinium enters the SnTe host lattice as Gd^{3+} ions, replacing the Sn^{2+} ions, and compensates the originally *p*-type material. The carrier concentrations and mobilities in all samples have been determined by Hall and magnetoresistance measurements. The Hall mobilities were of an order of 100 cm^2 V^{-1} s^{-1} from 77 K down to 4.2 K, and the hole concentration decreased nearly linearly with increasing Gd content. The hole-concentration values at 77 K are listed in Table I.

Magnetization measurements from 0.001 to 5.5 T were carried out using a superconducting quantum interference device (SQUID) magnetometer system. In order to determine the magnetic susceptibility the measurements were carried out over a temperature range from 5 to 300 K at four fields, usually between 0.001 and 0.05 T. The susceptibility and field offset at each temperature were determined by a linear-least-squares fit. The magnetization at fields from 0.01 to 5.5 T was measured over a temperature range from 2 to 25 K. The temperature varied by less than 0.01 K below 100 K and by less than 0.2 K for the highest measured temperatures. The high-field magnetization experiments at 4.2 K were carried out by the sample extraction method in steady magnetic fields up to 23 T in the High-Field Laboratory for Superconducting Materials at Tohoku University. Both water-cooled copper magnets, which produced magnetic fields up to 15 T, and hybrid magnets, which were composed of a superconducting magnet outside a water-cooled copper magnet and were capable of producing fields up to 31 T, were used. The magnetic field was determined with an accuracy of 2% and the errors in magnetization were typically 3%. The experimental details have been described previously.[8]

In order to interpret the magnetization measurements it was necessary to know the diamagnetic contribution of the host material χ_0. The susceptibility of single crystals of SnTe with hole concentrations from 1.5×10^{21} cm^{-3} to 1.8×10^{21} cm^{-3} was measured at temperatures from 2 to 150 K in magnetic fields from 0.5 to 2 T. The susceptibility was negative and showed some temperature dependence similar to that observed by Tovstyuk *et al.*[20] and Baginskij *et al.*[21] The data at temperatures above 25 K yielded χ_0 values from -3.6×10^{-7} to -5.6×10^{-7} emu/g for different samples. The temperature dependence may be explained by a diamagnetic free-carrier contribution to the susceptibility.[20]

It is very difficult to determine properly the contribution from the host lattice to the susceptibility of the $Sn_{1-x}Gd_xTe$ crystals, since χ_0 is carrier-concentration dependent[21] and in our materials the hole concentration changed with changing Gd content over an order of magnitude. Thus, the parameter χ_0 in $Sn_{1-x}Gd_xTe$ may be different from that in SnTe. From the analysis of our data for $Sn_{1-x}Gd_xTe$ and SnTe we found that the diamagnetic susceptibility of the lattice in magnetic fields above 0.1 T may be described well by the parameter $\chi_0=-5\times10^{-7}$ emu/g. This value also fits the low-field susceptibility data in samples with hole concentrations below 6×10^{20} cm^{-3}. Only in two samples with the highest hole concentration the low-field, high-temperature data indicated a higher absolute value of χ_0, in agreement with the results of Baginskij *et al.*[21] Therefore, in the analysis of the low-field susceptibility of these two samples, χ_0 was a fitting parameter. The χ_0 parameter values are given in Table I.

III. RESULTS AND DISCUSSION

The inverse susceptibility determined from the low-field magnetization measurements is shown in Fig. 1. The measurements were carried out up to 300 K in all samples. However, in the samples with low Gd content the magnetization was very small and changed sign with increasing temperature, due to the relatively high di-

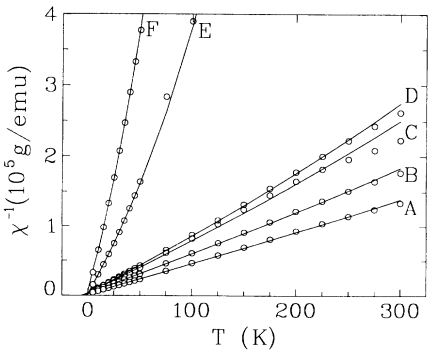

FIG. 1. Inverse susceptibility of $Sn_{1-x}Gd_xTe$. The circles represent the data and the solid lines were obtained from fits according to the Curie-Weiss law (see Table I).

amagnetic contribution from the host lattice.

The data have been fitted with a Curie-Weiss expression of the form

$$\chi=\frac{P_1}{T+\Theta}+\chi_0 , \tag{1}$$

where T is the absolute temperature and P_1 and Θ are fitting parameters. The parameter Θ is the Curie-Weiss temperature. Using the parameter P_1, we may obtain the effective number of magnetic ion spins $\bar{x}$ from the expression

$$\bar{x}=\frac{m_A+m_B}{S(S+1)(g_M\mu_B)^2N_A/(3k_BP_1)+m_A-m_M} , \tag{2}$$

where m_A, m_B, and m_M are the atomic masses of the cation, anion, and magnetic ion, respectively, N_A is the Avogadro number, S is the magnetic-ion spin, g_M is the magnetic-ion g factor, μ_B is the Bohr magneton, and k_B is the Boltzmann constant. For Gd^{3+} we take $g_M=2$ and $S=\frac{7}{2}$, as for Eu^{2+}. The data were fitted over different temperature ranges, because in the temperature region in which χ changed sign the signals were too small to obtain precise values of the susceptibility. The parameters $\bar{x}$ and Θ were determined with estimated errors of 10% and 20%, respectively. The parameter values are given in Table I.

Since the Curie-Weiss temperature Θ was much less than the measurement temperature, we could use this parameter to estimate the nearest-neighbor exchange interaction from the relation

$$\frac{J}{k_B}=-\frac{3\Theta}{2\bar{x}S(S+1)z} , \tag{3}$$

where $z=12$ is the number of nearest neighbors on cation sites. Since in Eq. (1) the denominator contains a plus sign, positive values of Θ and negative values of J/k_B imply an antiferromagnetic exchange. The values of J/k_B are shown in Table I. The average value of J/k_B is about -0.7 ± 0.2 K.

Magnetization as a function of magnetic field in fields up to 5.5 T, at temperatures from 2 to 25 K is shown in Figs. 2 and 3 for two samples with the highest and the lowest Gd content. The magnetization was fitted to the expression

$$M=M_s+\chi_0H , \tag{4}$$

where

$$M_S=M_0S\bar{x}_1B_s(\zeta) , \tag{5}$$

and

$$M_0=g_M\mu_BN_0 . \tag{6}$$

$B_s(\zeta)$ is a modified Brillouin function,

$$B_s(\zeta)=\frac{2S+1}{2S}\coth\left[\frac{2S+1}{2S}\zeta\right]-\frac{1}{2S}\coth\left[\frac{\zeta}{2S}\right] , \tag{7}$$

and

$$\zeta=\frac{Sg_M\mu_BH}{k_B(T+T_0)} . \tag{8}$$

The magnetization is given in emu/g, H is the magnetic field, N_0 is the number of cation sites per gram, and $\bar{x}_1$ is the effective number of isolated Gd^{3+} ions in cation sites, corresponding to $\bar{x}$ in the susceptibility fits. Here T_0 and $\bar{x}_1$ were fitting parameters, determined with an accuracy of about 10% and 20%, respectively. The solid lines in Figs. 2 and 3 are given by Eq. (4). The fitting parameters are listed in Table II. Such analyses were performed for all samples and the results were qualitatively similar to those presented in Figs. 2 and 3 and Table II. Results for the sample with $x_v=0.05$ (sample D) have been shown in Ref. 10.

In Fig. 2, which shows the magnetization for our highest Gd content, Eq. (4) does not fit well the magnetization data for temperatures below 5 K. Even for our lowest Gd content the fit is not very good at 2 K (Fig. 3). Moreover, the parameters obtained from fits at highest temperature are not reliable, since the Brillouin function loses its characteristic shape and becomes nearly a straight line. It was not possible to describe well the magnetization data at all temperatures by one set of parameters, as in $Pb_{1-x}Eu_xTe$,[9] although the parameters $\bar{x}_1$ and T_0 determined at 5 K agree fairly well with $\bar{x}$ and Θ obtained from the susceptibility data.

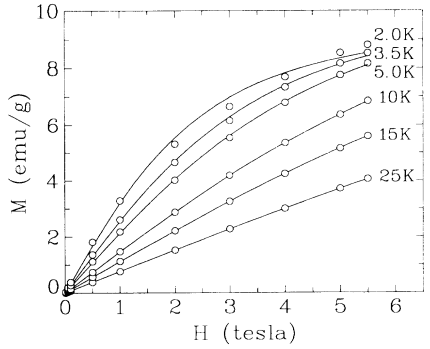

FIG. 2. Magnetization at temperatures up to 25 K of $Sn_{1-x}Gd_xTe$ with $x_v=0.09$, sample A. The circles represent the data and the solid lines were obtained from fits to the modified Brillouin function (see Table II).

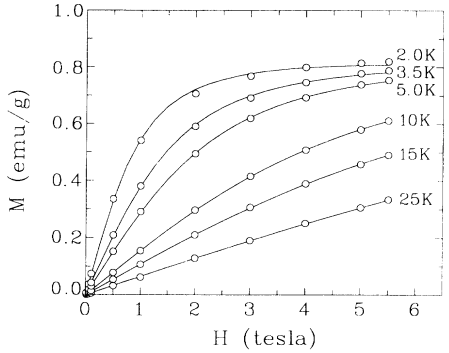

FIG. 3. Magnetization at temperatures up to 25 K of $Sn_{1-x}Gd_xTe$ with $x_v=0.006$, sample F. The circles represent the data and the solid lines were obtained from fits to the modified Brillouin function (see Table II).

As we have pointed out in our earlier paper, in order to describe properly the magnetization in IV-VI DMS we must take into account explicitly the pair-exchange interaction.[8] This approach gives good results when the measurements are carried out in magnetic fields high enough and at temperature low enough, that the magnetization nearly saturates. Therefore, we have measured the magnetization at fields up to 23 T, at 4.2 K, for all samples. The experimental data are shown in Fig. 4 as circles. The magnetization was now fitted with the expression

$$M = M_S + M_P + \chi_0 H \ , \tag{9}$$

where M_P is given as

$$M_P = \tfrac{1}{2} M_0 \bar{x}_2 \frac{\sum_{s=0}^{S_{\max}} \exp\left[\frac{J_P}{k_B T} s(s+1)\right] \sinh\left[\frac{2s+1}{2s}\xi_P\right] s B_s(\xi_P)}{\sum_{s=0}^{S_{\max}} \exp\left[\frac{J_P}{k_B T} s(s+1)\right] \sinh\left[\frac{2s+1}{2s}\xi_P\right]} \ , \tag{10}$$

where $\xi_P = s g_M \mu_B H / k_B T$, $S_{\max} = 2S$, $\bar{x}_2$ is the effective number of magnetic ions in pairs, and J_P is the pair-exchange parameter. This equation for M_P is essentially the same as that given by Bastard and Lewiner.[22] The term M_P yields a steplike behavior for $J_P/k_B T \gg 1$ and is similar to the Brillouin function for $J_P/k_B T < 1$.

Since from the susceptibility data we obtained $J/k_B < 1$ K, we did not expect to see steps in magnetization at $T = 4.2$ K, and indeed we do not see them in Fig. 4. However, though steps are not apparent in the data, magnetization versus magnetic field in the high-field region may be well described only when the pair-interaction function is included. First, we fitted our data to Eq. (9) with three fitting parameters: $\bar{x}_1$ in M_S, $\bar{x}_2$, and J_P in M_P. We fixed the parameter T_0 in M_S at 0, as we did previously for $Pb_{1-x}Gd_xTe$ and $Pb_{1-x}Eu_xTe$.[8,9] The results of the fits are shown in Fig. 4 as solid lines.

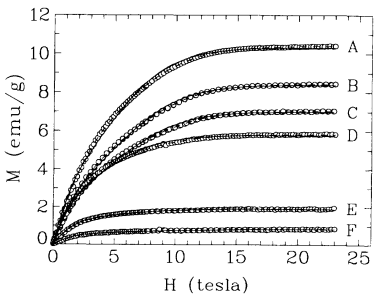

FIG. 4. High-field magnetization of $Sn_{1-x}Gd_xTe$ at 4.2 K. The circles represent the data. The solid lines were obtained from three-parameter fits, and the dashed lines were obtained from four-parameter fits (see Table III).

TABLE II. Brillouin-function fitting parameters for $Sn_{1-x}Gd_xTe$ for magnetization at T from 2 to 25 K for $H \leq 5.5$ T.

Sample	A		F	
T (K)	$\bar{x}_1$	T_0 (K)	$\bar{x}_1$	T_0 (K)
2	0.061	3.70	0.0054	0.36
2.5	0.063	3.97	0.0054	0.45
3	0.064	4.22	0.0054	0.45
3.5	0.065	4.36	0.0054	0.52
5	0.070	5.14	0.0055	0.65
10	0.080	7.18	0.0058	1.37
15	0.075	6.00	0.0059	1.45
25[a]	0.093	13.33	0.0068	6.38

[a]The fitting parameters were not reliable.

We see that reasonable fits were obtained. Since our data were sufficiently noise-free and close to saturation at highest fields, we were able to allow T_0 to vary and obtain fits with all four parameters, $\bar{x}_1$, T_0, $\bar{x}_2$, and J_P. In this case T_0 should represent the contribution to exchange from clusters other than pairs. The results are shown in Fig. 4 as dashed lines. We see that the four-parameter fits are even better than the three-parameter fits, though the difference is small.

A summary of the fitting parameters for high-magnetic-field data is given in Table III. The errors in parameters are about 15% for $\bar{x}_1$ and $\bar{x}_2$, 20% for T_0, and 30% for J_P/k_B. The parameters obtained from the three- and four-parameter fits are compatible and the exchange interaction estimated from T_0 in the four-parameter fits is considerably smaller that J_P/k_B. (We do not think, however, that the four-parameter fits should be trusted to obtain quantitative values of the exchange related to clusters other than pairs.) The average value of J_P/k_B is about -0.95 ± 0.15 K for three-parameter fits and -1.15 ± 0.25 K for four parameter fits, excluding sample F, where there may be not enough Gd pairs to obtain reliable values of the pair-exchange parameter. The values of J_P/k_B obtained from the three-parameter fits are slightly smaller than those obtained from the four-parameter fits, since in the three-parameter fits we obtain an "average" of the pair exchange and other smaller antiferromagnetic exchange parameters, while in the four-parameter fits the pair exchange is separated out. As we mentioned previously, the four-parameter fits are better in general; however, in samples with the lowest Gd content there was relatively more noise, and then the three-parameter fits seemed to be more reliable. This was also the case in our previous

TABLE III. Fitting parameters for $Sn_{1-x}Gd_xTe$ for high-field magnetization at 4.2 K.

Sample	$\bar{x}_v$	$\bar{x}_1$	$\bar{x}_2$	J_P/k_B (K)	T_0 (K)
A	0.09	0.026	0.040	−0.87	0
		0.039	0.028	−1.02	2.36
B	0.08	0.020	0.034	−0.95	0
		0.027	0.027	−1.06	1.77
C	0.06	0.019	0.026	−0.96	0
		0.025	0.020	−1.09	1.72
D	0.05	0.021	0.017	−0.80	0
		0.029	0.009	−1.08	1.85
E	0.01	0.010	0.003	−1.09	0
		0.011	0.002	−1.49	0.7
F	0.006	0.004	0.002	−1.69	0
		0.004	0.002	−2.21	0.9

analysis of $Pb_{1-x}Gd_xTe$ and other IV-VI DMS. Therefore, when comparing parameters obtained for different materials, we will refer to the results of the three-parameter fits. It should also be noted that Eq. (9) would describe the low-field data shown in Figs. 2 and 3 better than the modified Brillouin function, although these data alone are not sufficient to perform reliable three- and four-parameter fits.

A summary of the exchange parameter values for three rare-earth-doped IV-VI DMS is given in Table IV. We estimate that the errors in parameters are no more than 30%. The nearest-neighbor exchange interaction in $Sn_{1-x}Gd_xTe$ is antiferromagnetic, with the value of J_P/k_B of about −0.95 K. This is bigger in magnitude than −0.45 K in $Pb_{1-x}Gd_xTe$ (Ref. 8) and −0.46 K in $Pb_{1-x}Eu_xTe$.[9] This result is consistent with the model that bases the exchange interaction in II-VI and IV-VI DMS mainly on superexchange via the group-VI anion.[23] In this model the parameter J/k_B for the rare-earth ions is roughly proportional to d^{-20}, where d is the cation-anion separation. Since the lattice constants of SnTe and PbTe are 6.33 and 6.46 Å, respectively, the exchange interaction in $Sn_{1-x}Gd_xTe$ should be bigger than in $Pb_{1-x}Gd_xTe$ by a factor of about 1.5. Our data agree with this result, within the uncertainties in the parameter estimation.

TABLE IV. Exchange parameters in rare-earth doped IV-VI DMS.

Material	J/k_B (K)[a]	J/k_B (K)[b]
$Pb_{1-x}Eu_xTe$	−0.33	−0.46
$Pb_{1-x}Gd_xTe$	−0.36	−0.45
$Sn_{1-x}Gd_xTe$	−0.70	−0.95

[a]Susceptibility, Curie-Weiss-law fits.
[b]High-field magnetization, three-parameter fits.

The exchange-parameter values obtained from the fits of the susceptibility data to the Curie-Weiss law are slightly lower in magnitude than those obtained from fits of the high-field magnetization data to the full expression (9). A similar effect was observed in $Pb_{1-x}Gd_xTe$ (Refs. 7 and 8) and $Pb_{1-x}Eu_xTe$.[9] As was explained in Ref. 9, the exchange-parameter value determined from the Curie-Weiss temperature, is an average of all the exchange interactions among magnetic ions in the sample,[24] while the parameter J_P/k_B determined from the magnetization measurements in the high-field region depends mostly on the antiferromagnetic pair-exchange interaction. Therefore, it seems that comparison of the susceptibility and high-field data suggests the presence of a small ferromagnetic interaction, competitive with the main antiferromagnetic interaction.

Bruno *et al.* proposed for $Pb_{1-x}Gd_xTe$ a model, in which the nearest-neighbor exchange is negligible and the main exchange interaction is the next-nearest-neighbor exchange.[5] In this model our value of J/k_B would be doubled, since there are only six next-nearest neighbors. The suggestion is based on studies of the rare-earth chalcogenides, EuTe, GdS, and GdSe, with the rocksalt structure. In the Eu chalcogenides the nearest-neighbor exchange is claimed to be ferromagnetic,[25] and in the Gd chalcogenides to be antiferromagnetic,[26] while the next-nearest-neighbor exchange is antiferromagnetic in all rare-earth chalcogenides, except EuS; in the Gd compounds the next-nearest-neighbor interaction parameter J_2 is claimed to be bigger than the nearest-neighbor interaction parameter J_1.

It is not clear that the conclusions based on ordered magnetic compounds can be carried over to diluted rare-earth-doped semiconductors, in which no evidence of magnetic ordering has been found. Moreover, even in

rare-earth chalcogenides it is not a general rule that the nearest-neighbors are magnetically indifferent. In all the Eu compounds, except EuTe, the nearest-neighbor interaction dominates, and the relatively large values of the J_2 parameter in the metalliclike Gd compounds are attributed to the RKKY-type interaction. Also, Anderson has shown in a semiempirical analysis that, in the magnetic materials with the rocksalt structure, the 90° nearest-neighbor superexchange interaction may be antiferromagnetic and comparable to the 180° superexchange interaction. The exchange values depend on many factors, such as the symmetry of the interacting orbitals and the covalency of the cation-ligand bonds.[27]

Therefore, for our magnetically disordered, rare-earth-doped IV-VI DMS we use the same superexchange model as for Mn-doped IV-VI and II-VI DMS,[23] assuming that the magnetic properties of DMS are determined mainly by the nearest-neighbor superexchange interaction via an anion.

The exchange parameters J/k_B in rare-earth-doped DMS are smaller in magnitude than in $Pb_{1-x}Mn_xTe$ and other Mn-doped IV-VI DMS. The exchange interaction is expected to be smaller in DMS containing rare-earth ions than in those containing Mn, since the magnetic properties of rare-earth ions depend mostly on their f-shell electrons, which are shielded and bound more closely to the nucleus than the d-shell electrons in Mn. In our earlier paper we made some estimations of the exchange parameters in Mn-doped and rare-earth-doped DMS.[23] For rare-earth-doped DMS we calculated parameters J/k_B more than an order of magnitude lower than those actually observed. Kasuya, who encountered this problem in the europium chalcogenides, suggested that the superexchange interaction between the rare-earth ions includes an intra-atomic f-d interaction and an interatomic p-d interaction.[28] Since the latter is much stronger than the f-p exchange, Kasuya estimated that such a combined f-d and p-d mechanism would result in a correct magnitude of the exchange interaction in rare-earth-doped chalcogenides. This interesting suggestion needs further elaboration for application to rare-earth DMS systems.

An important difference between $Sn_{1-x}Gd_xTe$ and Pb-based IV-VI DMS is the high free-carrier concentration in the Sn-based chalcogenides. The hole concentration in $Sn_{1-x}Gd_xTe$ investigated here is above 10^{20} cm^{-3}, reaching 1.3×10^{21} cm^{-3} in samples with the lowest Gd content. In IV-VI Mn-doped DMS with similar carrier concentrations a carrier-induced magnetic ordering was observed. Mathur *et al.* reported a ferromagnetic ordering in $Sn_{1-x}Mn_xTe$.[2,11] Mauger and Escorne observed a ferromagnetic and reentrant spin-glass phase in $Sn_{1-x}Mn_xTe$.[12] Swagten *et al.*[13,14] and de Jonge *et al.*[15] also described a ferromagnetic and reentrant spin-glass phase in $Sn_{1-x}Mn_xTe$, though recent results of neutron-diffraction experiments reported by Vennix *et al.*[29] confirmed only the existence of a ferromagnetic phase. Hedgcock *et al.* observed a ferromagnetic ordering in $Sn_{1-x}Mn_xTe$ and $Pb_{1-x-y}Sn_yMn_xTe$,[16] and Story *et al.* found in $Pb_{1-x-y}Sn_yMn_xTe$ a reversible, carrier-induced transition between paramagnetic and ferromagnetic phases, at a hole concentration $p=3\times10^{20}$ cm^{-3}.[17,18] In $Sn_{0.97-x}Eu_xTe$ Mathur *et al.* did not report any magnetic ordering; however, their susceptibility data for $x\geq0.05$ showed a deviation from Curie-Weiss behavior similar to that in spin-glass materials.[2]

In our $Sn_{1-x}Gd_xTe$ samples we did not see any evidence of a ferromagnetic or spin-glass phase, at temperatures down to 2 K. In Figs. 1–3 we see that the temperature dependence of susceptibility and magnetic-field dependence of magnetization are the same as in conventional paramagnetic materials. The apparent lack of a RKKY-type exchange in $Sn_{1-x}Gd_xTe$ may be due to two reasons. First, Urban and Sperlich determined by electron-spin-resonance (ESR) measurements the exchange interaction between free carriers and magnetic ions in SnTe doped with Mn, Eu, and Gd and found that this interaction is about five times smaller for Gd ions than for Mn ions, and even less for Eu ions.[30] Thus, the RKKY interaction in the rare-earth-doped DMS may be considerably smaller than in Mn-doped DMS. Second, as was mentioned above, Gd atoms compensate the p-type SnTe and the hole concentration in $Sn_{1-x}Gd_xTe$ is nearly inversely proportional to the Gd content in this material. Therefore, when the carrier concentration is sufficient to induce long-range magnetic ordering, the Gd content may be too small, and vice versa.

IV. CONCLUSIONS

The magnetic properties of $Pb_{1-x}Eu_xTe$, $Pb_{1-x}Gd_xTe$, and $Sn_{1-x}Gd_xTe$ are similar. The average exchange interaction between the rare-earth ions is antiferromagnetic and the values of J/k_B lie below 1 K and are smaller than in manganese-doped, PbTe-based diluted magnetic semiconductors. This is probably due to the fact that the magnetic properties of rare-earth ions depend mostly on their f-shell electrons, which are shielded and bound more closely to the nucleus than the d-shell electrons in manganese.

In all three rare-earth-doped IV-VI chalcogenides a comparison of the high-field magnetization data and the low-field susceptibility data indicates that there may be a ferromagnetic contribution to the exchange, competitive with the main antiferromagnetic exchange, possibly from the next-nearest or more distant neighbors.

The exchange-interaction parameter J/k_B is larger in $Sn_{1-x}Gd_xTe$ than in $Pb_{1-x}Eu_xTe$ and $Pb_{1-x}Gd_xTe$. This result is consistent with the expectations of the model of superexchange interaction via anions in these materials, since the cation-anion spacing in SnTe is smaller than in PbTe.

In our experimental range of temperatures and magnetic fields we found no evidence for ferromagnetic or spin-glass ordering in $Sn_{1-x}Gd_xTe$. The lack of a long-range interaction may be explained by two facts: the exchange interaction between free carriers and magnetic ions in rare-earth doped SnTe is weak, and the Gd ions compensate the SnTe host material so that the hole concentration decreases with increasing Gd content. Thus, it may be difficult to obtain $Sn_{1-x}Gd_xTe$ in which both the carrier concentration and the Gd content would be sufficient to observe RKKY-type exchange interactions.

It is possible that $Sn_{1-x}Eu_xTe$ would show a long-range interaction, since the Eu ions would not be expected to compensate the SnTe host.

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