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The magnetic contribution to the specific heat of $Pb_{1-x}Gd_xTe$

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Abstract
The temperature dependence of the magnetic specific heat of the semimagnetic semiconductor $Pb_{1-x}Gd_xTe$ for $x = 0.033$ and 0.054, over the temperature range from 0.5 to 10 K, in magnetic fields up to 2 T, has been measured and analyzed theoretically. The maxima of the magnetic specific heat in nonzero magnetic fields are shifted to lower temperatures in comparison with theoretical predictions based on the simple cluster model. We propose a mechanism which explains these shifts. According to our model, they are caused by strong hybridization of the 5d shell of gadolinium with band states of the crystal.

1. Introduction

Semimagnetic semiconductors (SMSs), also known as diluted magnetic semiconductors (DMS), exhibit a number of interesting magnetic properties, which may lead to possible applications in spintronics (for a review see [1–4]). Our previous investigations of magnetic susceptibility and magnetization in Mn-doped and rare-earth-doped $A^{IV}B^{VI}$ SMSs have shown that the $A^{IV}B^{VI}$ SMSs with rare earths have a weaker exchange interaction than that found in the same materials with manganese as the magnetic ion [5–8]. These results have been well explained by the model of superexchange interaction between the nearest neighbors (NN) via an anion. Bindilatti *et al* and ter Haar *et al* investigated the high-field magnetization in $A^{IV}B^{VI}$ SMSs and found exchange constant values similar to ours [10, 9, 11]. A review of magnetic susceptibility and magnetization studies, including exchange among more distant magnetic ions, is given in [12].

The properties of magnetic ions in $A^{IV}B^{VI}$ SMSs and their exchange interactions are still not well understood. To develop a more complete model we performed complementary measurements of the magnetic specific heat of $Pb_{1-x}Mn_xTe$ and $Pb_{1-x}Eu_xTe$ [13, 14]. By the magnetic specific heat we mean the contribution to the specific heat due to the presence of magnetic ions, even in the absence of an external magnetic field. In order to explain the magnetic specific heat data we needed to take into account a splitting of the ground energy state of the single Eu ion in $Pb_{1-x}Eu_xTe$ and interaction of Mn ions with the free carriers in $Pb_{1-x}Mn_xTe$. In [14] we have also shown that the magnetic specific heat measurements reveal properties of the system (density of states) which are not reflected in the magnetization measurements.

In the present paper we examine the magnetic specific heat of another $A^{IV}B^{VI}$ rare-earth-doped crystal— $Pb_{1-x}Gd_xTe$. We measured the temperature dependence of two samples of $Pb_{1-x}Gd_xTe$ crystals with different Gd content. Some preliminary data for one sample have been previously reported [15]. In section 2 we present the experimental data and point out interesting differences with the data previously obtained for $Pb_{1-x}Mn_xTe$ and $Pb_{1-x}Eu_xTe$. In section 3 we propose a semiquantitative physical model to explain the results. According to our model, the results of magnetic specific heat measurements in $Pb_{1-x}Gd_xTe$ clearly show an influence of hybridization between 5d level of gadolinium and the band states of the crystal on the properties of an individual magnetic ion. The conclusions are presented in section 4.

2. Experiment

We have measured the specific heat of $Pb_{1-x}Gd_xTe$ with x-values of 0.033 and 0.054. The samples were prepared by

the Bridgman technique. The x-values were determined by microprobe and x-ray fluorescence analyses. We estimate the uncertainty in x-value to be about 20%. The samples were n-type with carrier concentrations of about 5.5×10^{19} cm^{-3} for $x = 0.033$ and 8.5×10^{19} cm^{-3} for $x = 0.054$. For the measurements we used crystals with a mass of about 7 mg, cleaved from larger boules.

Previously we measured high-temperature magnetic susceptibility and low-temperature, high-field magnetization of $Pb_{1-x}Gd_xTe$ with x up to 0.07 [5, 16]. By fitting the susceptibility data to the Curie–Weiss law we obtained the average Gd content in our samples (x_{av}) and very small Curie–Weiss temperatures indicating an antiferromagnetic exchange between Gd ions, J/k_B, of about -0.36 K.

The measurements of the heat capacity were performed using the thermal relaxation technique in a Quantum Design Physical Property Measurement System (PPMS) with the Helium-3 option. The temperature range was 0.4–20 K and the applied magnetic fields 0, 0.5, 1, and 2 T. The field was oriented at an arbitrary angle to the crystal.

In order to obtain the magnetic contribution to the specific heat, C_H, it was necessary to subtract the specific heat of the $Pb_{1-x}Gd_xTe$ lattice from the measured total specific heat of $Pb_{1-x}Gd_xTe$. This was not a simple process. Bevolo *et al* found that the specific heat of PbTe has an anomaly below 5 K and could not be fitted with the standard expression $C = \gamma T + \alpha T^3$, where γT and αT^3 are the electronic and lattice contributions, respectively [17]. In fact, they could not obtain a satisfactory fit to their data with an expression of the form, $C = \gamma T + \alpha T^3 + \sum_{i=1}^{n} \delta_i T^{2i+3}$ unless n was at least 10. Therefore, we measured the heat capacity of our own Bridgman-grown PbTe sample in zero magnetic field and 2 T, over the temperature range from 0.4 to 20 K and found that the temperature dependence was the same for 0 and 2 T within our experimental error. We also must take into account the effect that the replacement of Pb, with an atomic mass of 207.2, by Gd, with an atomic mass of 157.25, leads to a decrease in heat capacity, even for small values of x. To account for this we divided the entire set of PbTe specific heat data by the empirically determined factor 1.02 (same for $x = 0.033$ and 0.054 within an experimental uncertainty) before subtracting from the $Pb_{1-x}Gd_xTe$. The factor was determined by assuming that at temperatures above 15 K, in the absence of an applied magnetic field, the magnetic contribution to the specific heat of $Pb_{1-x}Gd_xTe$ is negligible. Therefore, this division by 1.02 gave results for PbTe that were the same as those for $Pb_{1-x}Gd_xTe$ at 15 K. This routine was described in more detail in [13] and [14]. Since this is an empirical correction, we emphasize in the present work the data at temperatures below 5 K where the lattice specific heat is much smaller than the total specific heat. In the interesting region, below 2 K, the specific heat of PbTe was more than 3 orders of magnitude smaller than that of $Pb_{1-x}Gd_xTe$.

For $Pb_{1-x}Gd_xTe$ the nuclear magnetic level splitting, Δ/k_B, in the magnetic field of 2 T would be about 5 mK and the Schottky specific heat is proportional to $(\Delta/T)^2$; therefore we estimated that the nuclear contribution to the specific heat above 0.4 K is negligible. The electronic contribution to the total specific heat becomes important in metals with carrier concentrations much higher than in our semiconductors. In fact, in $Pb_{1-x}Gd_xTe$ the electronic contribution below 5 K was estimated to be about 3 order of magnitude less than the measured magnetic contribution, but it made difficult the analysis of data at higher temperatures. This was the second reason to limit our analysis to data below 5 K.

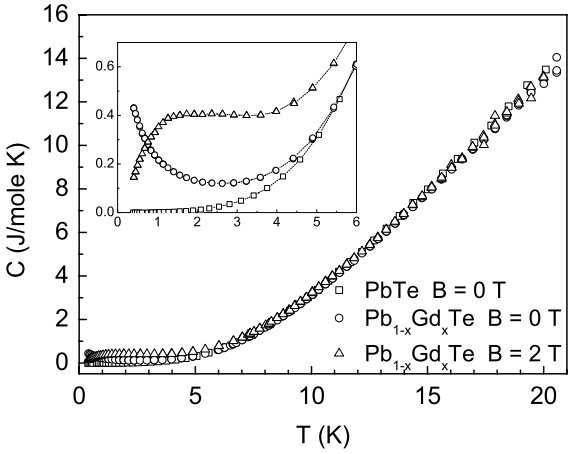

Figure 1. Molar specific heat of $Pb_{1-x}Gd_xTe$ with $x = 0.054$ and PbTe. Inset—the same data in the temperature region 0–6 K.

In figure 1 we show the total specific heat data of $Pb_{1-x}Gd_xTe$ before subtraction of the PbTe contribution together with the PbTe specific heat data before normalization. The inset with its expanded scale shows more clearly the different contribution to the heat capacity at low temperatures. The specific heats are plotted per mole of the compound.

The PPMS measurement errors in the region 0.4–5 K were below 1%. Taking into account the uncertainty in determining the molar specific heat of $Pb_{1-x}Gd_xTe$, PbTe, correction factor, and subtraction we estimate our experimental error in the specific heat to be about 3%.

$Pb_{1-x}Gd_xTe$ has a rock salt structure with a random distribution of magnetic ions in the cation sublattice. It does not have an easy magnetization axis. In our earlier magnetic susceptibility and magnetization measurements, as well as in our earlier and present specific heat measurements performed on samples with different magnetic field orientation with respect to the crystal axes we did not see any differences within our experimental error. We also estimated the geometrical corrections of the susceptibility that can modify the magnetic field within the material in our experiment. For that estimation we used our earlier magnetic susceptibility and magnetization data [18]. The demagnetization field at 0.4 K for the sample with $x = 0.054$ would change from 10 mT for an external magnetic field $B = 0.5$ T to 20 mT for an external magnetic field $B = 2$ T. For lower x and at higher temperatures it would be less. This is below 2% of the applied field, i.e. within our experimental error.

The magnetic specific heat experimental data for $Pb_{1-x}Gd_xTe$ are shown together with the predictions of the NN superexchange model in figure 2. The theoretical predictions were calculated with the exchange constant $J/k_B = -0.36$ K,

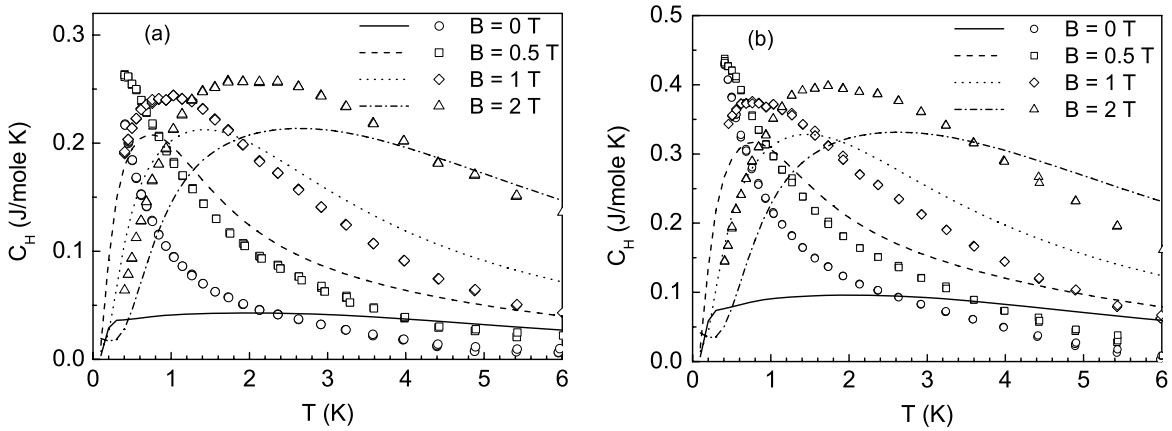

Figure 2. Magnetic specific heat of $Pb_{1-x}Gd_xTe$ in various magnetic fields. Points—experimental data, lines—theoretical predictions of the nearest neighbor interaction model. (a) $x = 0.033$, (b) $x = 0.054$.

which is based upon our experimental results mentioned above. The data for $B = 0$ and 0.5 T indicate a presence of a maximum in the specific heat below our experimental temperature range. We see that the apparent maximum at zero field is several times higher than that predicted by the model of NN superexchange. Similar behavior was observed in $Pb_{1-x}Mn_xTe$ and $Pb_{1-x}Eu_xTe$ [13, 14]. For $B = 1$ T we see a peak around 1 K and for $B = 2$ T a peak around 2 K. The height of the maximum is roughly independent of the magnetic field. This was also the case for $Pb_{1-x}Mn_xTe$, while for $Pb_{1-x}Eu_xTe$ peaks at zero field were noticeably smaller than those at 0.5 T and increased with increasing magnetic field. The specific heat of $Pb_{1-x}Gd_xTe$ looks qualitatively more similar to that of $Pb_{1-x}Mn_xTe$ than that of $Pb_{1-x}Eu_xTe$. This is surprising; we expected more similarities in PbTe with rare earth ions, Eu and Gd, for which the magnetic properties are related to the half filled f shell, while for $Pb_{1-x}Mn_xTe$ the Mn ion the magnetic properties are related to the half filled d shell. However, there is one specific difference between $Pb_{1-x}Gd_xTe$ and both $Pb_{1-x}Mn_xTe$ and $Pb_{1-x}Eu_xTe$. In $Pb_{1-x}Gd_xTe$ with $x = 0.054$ the maxima in the specific heat seem to occur at temperatures slightly lower than in $Pb_{1-x}Gd_xTe$ with $x = 0.033$. In both $Pb_{1-x}Mn_xTe$ and $Pb_{1-x}Eu_xTe$ the maxima in the specific heat shifted with increasing x to higher temperatures.

3. Theoretical analysis

We start our analysis of the magnetic specific heat of $Pb_{1-x}Gd_xTe$ from a comparison of the experimental results with the predictions of the NN superexchange model. In figure 2 we see at least two characteristic features.

First, the tails of the magnetic specific heat for $Pb_{1-x}Gd_xTe$ in all magnetic fields decay faster than the model predicts. In the absence of the magnetic field the magnetic specific heat drops close to zero at temperatures above 6 K. This is in strong contrast to the $Pb_{1-x}Eu_xTe$ case where the magnetic specific heat is nonzero for much higher temperatures. In our previous paper we proposed that the long tails in magnetic specific heat for $Pb_{1-x}Eu_xTe$ are due to large splitting of the ground state of Eu^{2+} ions due to the disordered crystal environment, and to some extent due to the presence of Eu–Eu pairs. In the present case the data for $B = 0$ suggest that the ground state splitting is much smaller and the Gd–Gd interaction is very small. This suggests that the main contribution to the specific heat comes from singles and actually this is the basic assumption in the following analysis.

Second, and what is in our opinion the most interesting feature, the peaks in the magnetic specific heat for $Pb_{1-x}Gd_xTe$ appear at lower temperatures than the theoretical predictions of the model of NN exchange interaction only. It means that the magnetic specific heat in nonzero magnetic field behaves as if it had been measured in lower magnetic field than the field actually applied in the experiment. Alternatively one may say that the g-factor for 4f electrons is much lower than 2. It turns out that the experimental data for nonzero magnetic field may be described very well assuming that the effective g-factor equals 1.6. However we do not see any explanation for such a value. That is why, below, we propose another mechanism which leads to additional 'artificial' magnetic field, the direction of which is opposite to the external magnetic field.

The magnetic specific heat measurements are not the only experiments showing important differences between Eu and Gd ions in the PbTe host lattice. In electron paramagnetic resonance one measures the parameter b_4 which describes interaction of the 4f shell electrons with the crystal environment [19]. From experiment it turns out that b_4 is positive for europium and negative for gadolinium. In [20] it was shown that different positions of 5d level of gadolinium and europium are responsible for the opposite signs of b_4. In the case of PbGdTe the 5d level of gadolinium lies about 0.2 eV above the conduction band minimum, while for Eu in PbEuTe the energy of the 5d level is about 2 eV higher.

We think that this difference is responsible for the large difference in the magnetic specific heat between $Pb_{1-x}Eu_xTe$ and $Pb_{1-x}Gd_xTe$. The considerations and formulae in [20] apply only to situations when the content of Gd in PbGdTe is very small. Then the Fermi energy lies below the conduction band and 5d level is unoccupied. If the content of Gd is higher, as in the present case, the position of the Fermi

energy level is higher and eventually this level lies in the conduction band. Indeed our samples were of n-type and the concentration of free electrons was between 5.5×10^{19} cm^{-3} and 8.5×10^{19} cm^{-3}. Notice that the number of free carriers per cubic centimeter is approximately an order of magnitude smaller than the number of gadolinium ions. According to considerations in [21], we expect that most of the gadolinium ions are in the Gd^{2+} state, i.e. with one electron in the 5d level. The electron on 5d shell interacts strongly with 4f electrons via an exchange interaction. This interaction together with hybridization between 5d states and atomic orbitals of the surrounding ions leads to a smaller 4f spin splitting in the applied magnetic field, which in turn leads to the displacement of the peak to lower temperatures. A simple semiquantitative model describing the mechanism is presented below.

Let us consider a free Gd^{2+} ion with seven electrons on its 4f shell and one electron on its 5d shell in a magnetic field B directed along the z axis. The Hamiltonian describing interaction between 4f and 5d electrons and interaction with the external magnetic field reads

$$H = -J_{\mathrm{fd}}\mathbf{S}\cdot\mathbf{s} + g\mu_{\mathrm{B}}B(S_z + s_z), \tag{1}$$

where $S = 7/2$ is the total spin of 4f electrons, $s = 1/2$ is the spin of 5d electron, S_z and s_z are the projections of the spins along the z axis, and J_{fd} is the exchange constant. From the optical data we know that J_{fd} is of the order of 0.25 eV [22]. In the external magnetic field the ion is polarized, and the average value $\langle S_z\rangle$ of zth component of the spin of the 4f electrons is nonzero. One may say that the electron on the 5d shell is in an effective magnetic field

$$h_{\mathrm{eff}} = B + \frac{J_{\mathrm{fd}}\langle S_z\rangle}{g\mu_{\mathrm{B}}}, \tag{2}$$

where the g-factor is assumed to be 2, which is generally a reasonable approximation for semiconductors, and μ_{B} is the Bohr magneton. The energy difference between spin up and spin down states of the 5d electron (spin splitting) is

$$\Delta E = g\mu_{\mathrm{B}}B + J_{\mathrm{fd}}\langle S_z\rangle \tag{3}$$

and in our magnetic field range is of the order of J_{fd}.

Now, if we put the ion into the crystal lattice the 5d states are hybridized with the orbitals of the surrounding atoms. This hybridization is strong because the 5d orbitals are extended in space and the overlap with the orbitals of the surrounding atoms is large. Let us, for simplicity of argument, neglect the spin orbit interaction in the band states of the host crystal. (This certainly cannot be done for PbTe in a more quantitative description). Then the spin up (spin down) 5d states hybridize with spin up (spin down) states of band states. Consequently the 5d spin splitting caused mainly by the second term in equation (3) via hybridization 'tries' to split band states. Such band states splitting leads to an increase of the kinetic energy of band electrons. On the other hand the system 'tends' to minimize the kinetic energy of band electrons. This tendency, via hybridization, results in a decrease of spin splitting of 5d states.

Let us summarize double role of hybridization between magnetic ion orbitals and the band states of the host crystal. On the one hand, provided the magnetic ion is polarized, the hybridization leads to spin splitting of band states. This is well known, the whole semimagnetic semiconductors physics is based on this fact. On the other hand, if our semiquantitative description proposed below is correct the results of the presented experiment for $Pb_{1-x}Gd_xTe$ provide a very interesting and a direct evidence of the reverse process. The hybridization leads to diminishing of 5d spin splitting and consequently, as will be shown below, to the significant change of 4f electron spin splitting.

To put the above qualitative considerations onto more quantitative level we propose that the quantity describing the 5d spin splitting is the average of the zth component of 5d spin $\langle s_z\rangle$. Such an assumption leads in a natural way to an internal magnetic field acting on the 5d spin. As has been explained above, due to system's tendency to minimize energy, hybridization puts certain restrictions on the 5d spin splitting. It means that all thermodynamic quantities for the Gd ion, in particular magnetic specific heat, should be calculated with an additional constraint, $\langle s_z\rangle = \mathrm{const}$, put on the average value of 5d spin. This constant, in general, depends on the external magnetic field and temperature. For simplicity, however, the temperature dependence will be neglected. Then, using the concept of Lagrange multipliers we add to the Hamiltonian, equation (1), a term proportional to s_z,

$$H = -J_{\mathrm{fd}}\mathbf{S}\cdot\mathbf{s} + g\mu_{\mathrm{B}}B(S_z + s_z) + g\mu_{\mathrm{B}}b_{\mathrm{art}}s_z, \tag{4}$$

where the Lagrange multiplier is written in the form $g\mu_{\mathrm{B}}b_{\mathrm{art}}$. Although this form is very suggestive, we stress that b_{art} is not the real magnetic field. Therefore we will call it the artificial magnetic field. The last term in equation (4) only describes the influence of surrounding ions on the 5d spin splitting. The external magnetic field dependent parameter b_{art} is treated as the only fitting parameter.

The comparison of the above approach with experimental results is presented in figure 3. We see that the experiment is quite well described assuming that the artificial magnetic field is proportional to the real, external magnetic field, $b_{\mathrm{art}} = -2.5B$, at least in the investigated range of temperatures and magnetic fields. We should note, however, that although the sign of b_{art} should be negative to explain our experimental results and its values should be approximately those shown in figure 3, we can not state explicitly that b_{art} is proportional to the applied field.

We realize that in the sample there are also gadolinium ions in the Gd^{3+} state. However, even assuming that 30% of the magnetic ions are in the Gd^{3+} state, which is much more than we expect based on the free carrier density and the Gd density, the final results, shown in figure 3 do not change appreciably. For simplicity, in the analysis we assume that all gadolinium ions are in Gd^{2+} state.

As in our previous works devoted to magnetic specific heat, in the present work the theoretical analysis concerns primarily the sample with the smallest magnetic ion content. This is because we believe that the magnetic specific heat is determined mainly by the properties of single magnetic ions. In

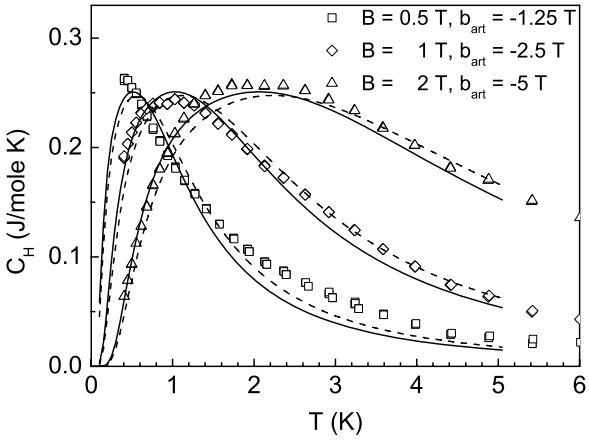

Figure 3. Magnetic specific heat for different magnetic fields in $Pb_{1-x}Gd_xTe$ with $x = 0.033$. Symbols—experimental results, lines—theoretical calculations for indicated artificial magnetic fields (b_{art}—see equation (4).). Solid lines calculated assuming all gadolinium ions are Gd^{2+}, dashed lines calculated assuming 30% gadolinium ions are Gd^{3+}.

samples with higher concentrations a more and more important role is played by ion–ion interactions and it is difficult to determine which effects are caused by single ions and which are caused by ion–ion interactions.

At the end of section 2 we mentioned that the peaks in magnetic specific heat in nonzero magnetic field for the sample with higher gadolinium concentration are at lower temperature than the peaks for the sample with lower Gd concentration. Notice that, qualitatively, our model is in agreement with this observation. Higher Gd concentration also means a higher free carriers concentration, higher Fermi energy, and higher occupancy of the 5d level. We expect then that the influence of hybridization on the 4f states via 5d level should be stronger in samples with a higher concentration of Gd. This results, according to our model, in a further decrease of 4f states splitting. For smaller 4f spin splitting the peak in magnetic specific heat moves to lower temperatures.

The picture proposed above is valid only for nonzero external magnetic field. For $B = 0$ the average value of the 5d spin, $\langle s_z \rangle$, must be equal to zero because of time reversal symmetry. Then, it is not sensible to put any constraint on $\langle s_z \rangle$ because demanding $\langle s_z \rangle \neq 0$ we would break this symmetry artificially. For $B = 0$, in our model, the only term in the Hamiltonian is the term $-J_{fd}\mathbf{S} \cdot \mathbf{s}$. This term contributes to the C_H at zero magnetic field, but is significant only at high temperatures of the order of several thousand Kelvins.

As noticed in section 2 the magnetic specific heat in the present case looks similar to that of $Pb_{1-x}Mn_xTe$ [13]. In [13] we concluded that the interaction between localized magnetic moments of magnetic ions and spins of free carriers was responsible for the high peak in zero external magnetic field. Unfortunately, the model from [13] cannot be applied directly in the present case, even with replacement of the manganese spin $S = 5/2$ by the gadolinium spin $S = 7/2$, because the interaction between free carriers and magnetic 4f shell is not direct but via 5d orbitals. That leads to much more complicated, difficult to solve, Hamiltonian. However, as we expect, the qualitative conclusions should be similar in both cases.

In the present paper we have concentrated on the behavior of magnetic specific heat in the presence of an external magnetic field. That is why our model is very simple and contains only the terms that are necessary to explain this behavior. The model does not take into account several factors which certainly influence the magnetic specific heat, particularly for $B = 0$, for example the spin–orbit interaction for the 5d electron or the influence of the crystal field on the 5d level. The more precise description of the magnetic specific heat in zero external magnetic field remains an open problem.

4. Conclusions

We have measured the magnetic specific heat of $Pb_{1-x}Gd_xTe$ for $x = 0.033$ and 0.054. The most interesting feature of the present experimental results in comparison to the previously measured $Pb_{1-x}Mn_xTe$ and $Pb_{1-x}Eu_xTe$ crystals, is the appearance of magnetic specific heat peak at temperatures lower than the nearest neighbor exchange model predicts. To the best of our knowledge this effect is new and has not been observed previously either in IV–VI or in II–VI semimagnetic semiconductors. In our opinion the hybridization between 5d level of gadolinium and crystal band states is responsible for this effect. The effect of this hybridization is enhanced in $Pb_{1-x}Gd_xTe$ because the energy of the gadolinium 5d level lies close to the bottom of the conduction band. The presented data and theoretical analysis show the importance of 5d level of rare earth ions in semimagnetic semiconductors. In particular, comparing the experimental data for $Pb_{1-x}Gd_xTe$ and $Pb_{1-x}Eu_xTe$ we see that the real, in the present case, and virtual, in the case of $Pb_{1-x}Eu_xTe$ occupancy leads to a quite different behavior of macroscopic quantities, like magnetic specific heat.

Acknowledgments

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