Magnetic contribution to the specific heat of $Pb_{1-x}Mn_xTe$

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(Received 22 October 2001; published 4 April 2002)

Temperature dependence of magnetic specific heat and magnetic susceptibility has been studied experimentally and theoretically in the semimagnetic semiconductor $Pb_{1-x}Mn_xTe$ for $x=0.024$ and $x=0.056$, over the temperature range from 0.5 to 15 K, in magnetic fields up to 4 T. There was usually a maximum in the magnetic specific heat around 1 K in zero and low magnetic fields; the maximum shifted toward higher temperatures with increasing magnetic field. The experimental data have been analyzed in the framework of magnetic cluster models. An analysis of the influence of local lattice distortions and *sp-d* exchange coupling has been performed.

DOI: 10.1103/PhysRevB.65.165206 PACS number(s): 71.70.Gm, 75.40.Cx

I. INTRODUCTION

Semimagnetic semiconductors (SMS's), also known as diluted magnetic semiconductors, are semiconducting alloys with substitutional magnetic ions. They exhibit a number of interesting properties like interesting magneto-optical effects related to the giant Zeeman splitting of band states,[1–4] carrier induced ferromagnetism,[5,6] and the formation of bound magnetic polarons.[7] The main origin of this rich variety of phenomena is the *sp-d* exchange interaction between delocalized band carriers and the localized magnetic moments introduced into the host lattice by the magnetic ions. The intensive studies of SMS's for more than two decades were devoted to SMS's based on II-VI or IV-VI semiconducting compounds with $Cd_{1-x}Mn_xTe$ and $Pb_{1-x}Mn_xTe$ as representative examples. Recently we observed considerable interest in SMS's based on III-V compounds, namely GaMnAs and InMnAs.[8] This is connected with the paramagnetic-ferromagnetic transition observed in these compounds at relatively high temperatures up to 110 K.

The present paper is devoted to experimental and theoretical studies of the magnetic specific heat of $Pb_{1-x}Mn_xTe$, a paramagnetic IV-VI semimagnetic semiconductor. Magnetization and magnetic susceptibility measurements have shown that, in general, IV-VI SMS's with a $3d$ element as the magnetic ion have a much weaker exchange interaction than that found in II-VI SMS's with the same magnetic ion. However, the mechanism of the exchange interaction among the magnetic ions is still not well understood. For example, because IV-VI chalcogenide systems crystallize in the rocksalt structure, one might suspect a more pronounced role for the next-nearest-neighbor (NNN) interaction[9] compared to II-VI materials, where this NNN interaction is usually neglected, at least in short-range-order interaction models.

Our main motivation for the present research was to develop a more complete model of the exchange interaction and to obtain parameters for this interaction among magnetic ions by taking into account the results of specific-heat studies together with the complementary results of magnetization and magnetic susceptibility measurements. Up to now, no experimental data on the magnetic contribution to the specific heat of $Pb_{1-x}Mn_xTe$ have been available. Therefore in this work we examine the temperature and magnetic-field dependence of the magnetic specific heat of two samples of $Pb_{1-x}Mn_xTe$ with different Mn content. The details of the experiment, our approach for extracting the magnetic part from the total measured heat capacity, and the results of the measurements are presented in Sec. II. In Sec. III we apply a number of theoretical models to explain the experimental data. Apart from the models already known in the literature, we analyze the effect of splitting of the ground state of manganese ion brought about by the *sp-d* interaction between localized magnetic moments and free carriers. Section IV contains the summary and conclusions.

II. EXPERIMENT

We have measured the specific heat of $Pb_{1-x}Mn_xTe$ with x values of 0.024 and 0.056. The samples were grown by the Bridgman technique. The Mn content in the samples was determined by x-ray fluorescent energy dispersive analysis with an accuracy of about 7% of the x value. As expected, the lattice constant of our $Pb_{1-x}Mn_xTe$ crystals decreased linearly with increasing Mn content according to Vegard's law, $a(x)=(6.46-0.632x)$ Å. X-ray powder diffraction analyses were also performed on our samples. We did not see any peaks corresponding to phases other than the rocksalt crystal of $Pb_{1-x}Mn_xTe$. Carrier concentrations and mobilities were determined by the Hall effect and conductivity measurements. All samples were p type. At 77 K the hole concentration both for $x=0.056$ and $x=0.024$ was about 3×10^{18} cm^{-3}, and the hole mobility was about 2.5×10^{3} $cm^2/V\,s$. For the measurements of the heat capacity we used samples in the form of disks with diameters up to 8 mm. Each sample consisted of several large monocrystalline grains.

Previous measurements of high-temperature magnetic susceptibility and low-temperature, high-field magnetization have shown that $Pb_{1-x}Mn_xTe$ with x up to 0.04 was paramagnetic with a weak antiferromagnetic exchange interaction between Mn ions.[10–12] We performed measurements of the magnetic susceptibility of our $Pb_{1-x}Mn_xTe$ samples with x up to 0.09. In Fig. 1 we show the inverse susceptibility, obtained from these measurements, vs temperature up to 80

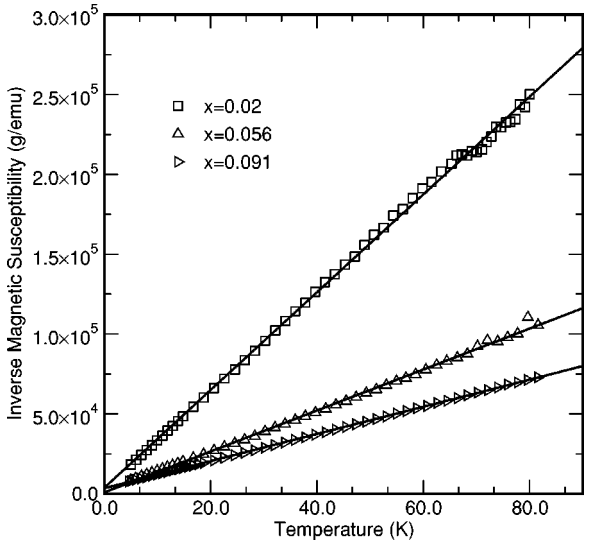

FIG. 1. Inverse magnetic susceptibility of $Pb_{1-x}Mn_xTe$ vs temperature for various Mn compositions.

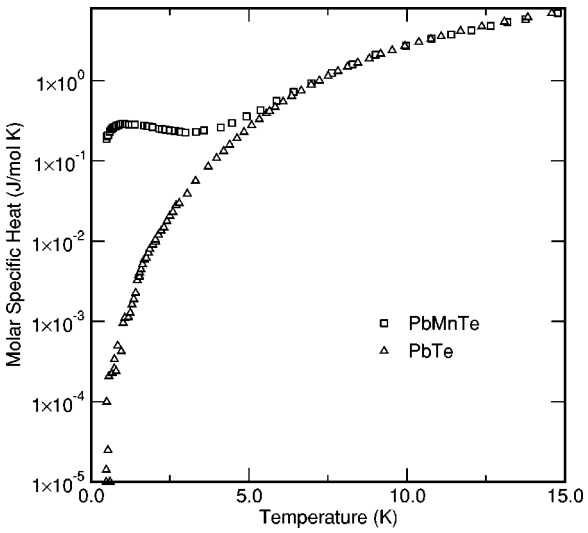

FIG. 2. Specific heat of $Pb_{1-x}Mn_xTe$ with $x=0.056$ (circles) and of PbTe (diamonds) in zero magnetic field.

K. These results confirmed the presence of this weak antiferromagnetic exchange interaction and agree well with literature data.[10–12]

The measurements of the heat capacity were performed in a cryostat using a ^{3}He or ^{4}He system, over the temperature range 0.5–15 K, in magnetic fields 0, 0.5, 2, and 4 T. We used the standard adiabatic heat-pulse method.[13] Errors in the heat-capacity values were about 5%. In order to determine the specific heat of the sample we needed first to subtract the heat capacity of the calorimeter from the total heat capacity. Since the heat capacity of the calorimeter was too small for the adiabatic method, we measured the heat capacity of the calorimeter in magnetic fields 0, 2, and 4 T using the relaxation method, developed by Bachman *et al.*[14] The specific heat of the calorimeter did not depend on the magnetic field, within our experimental error.

In order to obtain the magnetic contribution to the specific heat C_m, it was necessary to subtract the specific heat of the PbTe lattice from the measured specific heat of $Pb_{1-x}Mn_xTe$. This, it turned out, 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.[15] In fact, they could not obtain a satisfactory fit to their data with an expression of the form $C=\gamma T+\alpha T^3+\Sigma_{i=1}^{n}\delta_i T^{2i+3}$ unless n was at least 10. Therefore we measured the heat capacity of our own PbTe sample, which was also grown by the Bridgman method. At temperatures below 4 K the heat capacity of PbTe was very small and we found it necessary to measure by the relaxation method; above 4 K it was possible to use the same adiabatic method as we used for $Pb_{1-x}Mn_xTe$.

We measured the heat capacity of PbTe in 0 magnetic field and at 2 T over the temperature range from 0.5 to 15 K and, like the calorimeter, found that the temperature dependence was the same for 0 and 2 T within our experimental error. The specific heat of PbTe above 10 K was greater than that of $Pb_{1-x}Mn_xTe$, both with $x=0.056$ and $x=0.024$, due to the lattice contribution. That is, the replacement of Pb with an atomic mass of 207.2 by Mn with an atomic mass of 54.9 leads to a decrease in heat capacity, even for such small values of x. To take this effect into account we divided the entire set of PbTe specific-heat data by empirically determined factors, 1.05 for $x=0.056$ and 1.03 for $x=0.024$, before subtracting from the $Pb_{1-x}Mn_xTe$. These factors were 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 $Pb_{1-x}Mn_xTe$ is negligible. This division by 1.05 (1.03) gave results for PbTe that were the same as those for $Pb_{1-x}Mn_xTe$ at 15 K for $x=0.056$ (0.024). 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. The specific heat values for $Pb_{1-x}Mn_xTe$ with $x=0.056$, including the host lattice contribution of PbTe are shown in Fig. 2. In the interesting region, below 2 K, the specific heat of PbTe was more than 3 orders of magnitude smaller than that of $Pb_{1-x}Mn_xTe$.

The magnetic specific-heat data for $Pb_{1-x}Mn_xTe$ are shown in Figs. 3 and 4. We believe that the scatter in the data is the experimental error. For $x=0.056$ there is a maximum in the magnetic specific heat at about 1 K, in zero magnetic field. In higher magnetic fields the peak shifts to higher temperatures. The same field dependence of the magnetic specific heat is observed in $Pb_{1-x}Mn_xTe$ with $x=0.024$, but in this case the peak at $H=0.5$ T appears at a temperature lower than for $x=0.056$, and the peak in zero magnetic field is below our experimental temperature range. A similar behavior of the magnetic specific heat was observed previously in other IV-VI SMS's, such as $Sn_{1-x}Mn_xTe$.[16]

III. THEORETICAL MODELS

At first we apply the theoretical models developed for the description of magnetic specific heat in II-VI SMS's in a number of papers (see, for example, Refs. 17–19). In accor-

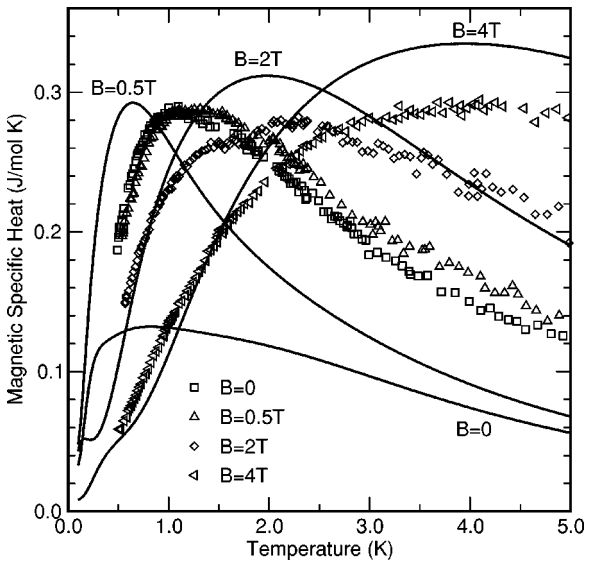

FIG. 3. Magnetic specific heat of $Pb_{1-x}Mn_xTe$ with $x=0.056$ in various magnetic fields. Points: experimental data; lines: theoretical predictions of NNN cluster model.

dance with the simplest random distribution model[17,18] (also called cluster model), we assumed that the magnetic spin-spin interactions are short range and the manganese ions are randomly distributed over the cation sites of the lattice. These assumptions are quite well justified in SMS's with small Mn content and fully supported by the earlier theoretical analysis of magnetic susceptibility and magnetization measurements.[11,20] For the given concentration of magnetic ions, we should find a certain number of singles, pairs, triples, and larger magnetic clusters as predicted statistically. Because the manganese concentration is rather small, in our analysis we have limited ourselves to singles, pairs, and triples configurations. This is particularly well justified for

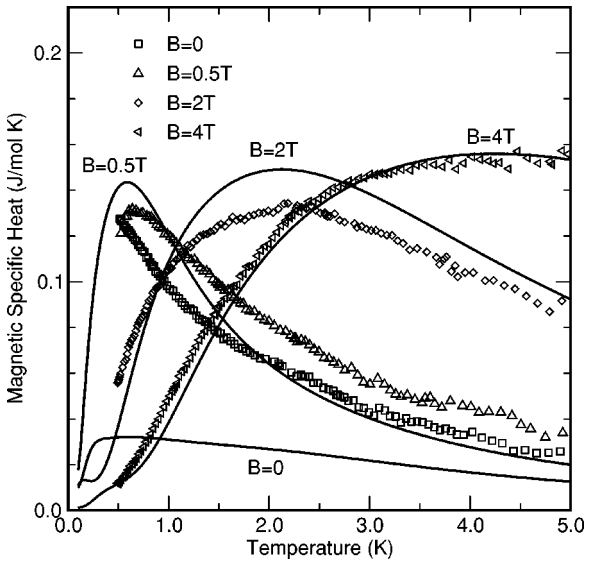

FIG. 4. Magnetic specific heat of $Pb_{1-x}Mn_xTe$ with $x=0.024$ in various magnetic fields. Points: experimental data, lines: theoretical predictions of NNN cluster model.

the 0.024 sample as the manganese atoms in these configurations constitute more than 98% of the magnetic atoms system in the case of magnetic interaction limited to nearest neighbors (NN). If the exchange interaction is extended to the next-nearest neighbors (NNN) then considering these configurations we take into account about 96% of Mn atoms. For the 0.056 sample the situation is more complex, because the percentage of Mn atoms in other configurations is much larger and, strictly speaking, the cluster model which takes into account singles, pairs, and triples only does not apply. For this sample we have calculated the numbers of singles, pairs, and triples according to statistical distribution. Next, in order to take into account Mn atoms in the remaining configurations, we have multiplied the numbers of pairs and triples by a properly chosen factor. We think that such a procedure enables us to draw at least semiquantitative conclusions. The interaction of manganese spins was described by the Heisenberg Hamiltonian with an additional Zeeman term due to an external magnetic field. The exchange integral for the nearest-neighbor interaction is known from high-temperature susceptibility and magnetization measurements and is of the order of 1 K with an antiferromagnetic sign.[11] Since for such simple configurations of exchange coupled spins the eigenvalue problem may be easily solved and for the fcc lattice the average numbers of singles, pairs, and triples are known too,[21] there is no difficulty in calculating the magnetic specific heat for the whole system.

The results of calculations are presented in Fig. 3 and Fig. 4 (solid lines). We adopted here the cluster model taking into account NN and NNN interactions with the exchange constants $J_{NN}=-0.8$ K and $J_{NNN}=-0.2$ K consistent with magnetic susceptibility measurements. One can see that the predictions of the cluster models reasonably well quantitatively agree with our experimental data for high magnetic fields and for low Mn content. The agreement of the theory and experiment becomes worse for lower fields and higher Mn compositions and breaks down completely for zero external magnetic field. This is the most striking experimental finding particularly clearly seen for the sample with 5.6% of Mn, for which the experimental curves taken at $B=0$ and $B=0.5$ T are almost identical. This qualitatively different feature of $C_H(T)$ dependence, not found in other cubic paramagnetic SMS materials with Mn, constitutes the main challenge of theoretical analysis of magnetic specific heat of PbMnTe. Below, we analyze the problem within the framework of different theoretical models. The calculated curves together with experimental data for $B=0$ are presented in Fig. 5.

Qualitatively, in the NN cluster model described above, the temperature position of the peak in the magnetic specific heat corresponds to the energy-level splitting of nearest-neighbor pairs (for simplicity of discussion we neglect triples). The height of the peak, on the other hand, reflects the number of such pairs contributing to the specific heat. In this respect, let us remind ourselves that in zero magnetic field the magnetic specific heat of singles is equal to zero, because, due to degeneracy of their energy levels, they cannot absorb energy. This degeneracy is lifted in an external

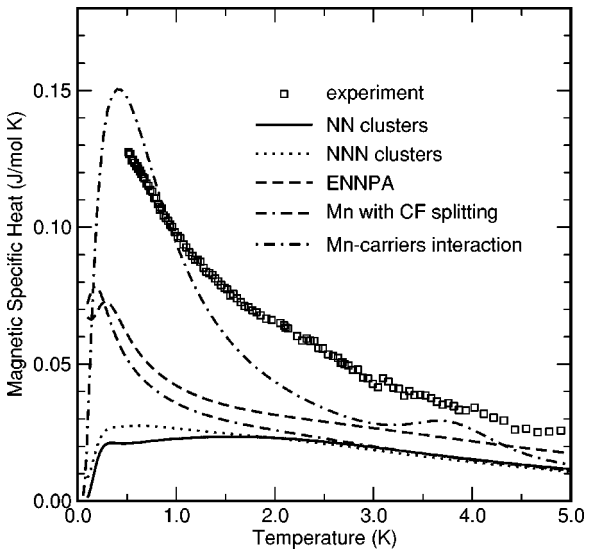

FIG. 5. Comparison of the magnetic specific heat of $Pb_{0.976}Mn_{0.024}Te$ calculated according to different models in zero external magnetic field with the experimental results.

magnetic field and only then do the singles contribute to the magnetic specific heat.

As we have already noticed, one of the most striking features of the experimental data is the equality of the peaks' heights for zero and nonzero magnetic field. This suggests that there are no singles in the system. In other words, a model based on short-range spin-spin interaction and a random distribution of the magnetic ions in the lattice cannot explain the experiment.

In order to learn about the distance dependence of the exchange integral between two magnetic ions in PbMnTe we have performed numerical calculations,[22] analogous to those performed in Ref. 23; superexchange and the Blombergen-Rowland mechanism were taken into account. The calculated exchange integral is of antiferromagnetic sign and decreases very quickly with the distance. The main contribution to the Curie-Weiss temperature θ comes from the first and second coordination zones. This precludes any assumption of an extraordinary long-range order of magnetic interactions.

Although from the numerical calculations point of view one should not expect long order interactions; for completeness, we tried to describe the behavior of the specific heat with the aid of the so-called extended nearest-neighbor pair approximation (ENNPA) model. The ENNPA was successfully applied in a number of papers to the description of the magnetization or magnetic specific-heat measurements (see, for example, Ref. 19). In the ENNPA model one assumes long ranged magnetic interactions with an exchange integral of the form $J(R)=J_{NN}(R/R_0)^{-\kappa}$. Here J_{NN} is a constant of the order of the exchange integral for nearest neighbors separated by a distance R_0 and κ is an exponent characterizing the decrease of the exchange integral with increasing distance R between two magnetic ions. We do not describe the model in detail because this has been done previously (see, for example, Refs. 19 and 24).

The ENNPA model (see Fig. 5, dashed line) provides a somewhat better description of the experiment than the simplest cluster models giving rise to smaller difference between the specific heat in zero and nonzero magnetic field. However, in our opinion, this difference is still too large to accept ENNPA as the description of the experiment. For the results presented in Fig. 5 it was assumed that $J_{NN}=-1$ K and $\kappa=4$. No significant improvement was achieved by any changes of these parameters within sensible limits.

We have done a number of other simulations which we do not describe here in detail. In these simulations larger clusters have been taken into account, different sets of exchange constants have been assumed, sometimes disorder has been included. None of these models provided satisfactory explanation of the experiment. Summarizing the above considerations, we conclude that the experimental results probably cannot be described by any model based on a Hamiltonian containing the interactions between manganese spins only.

Let us try now to look for other possible mechanisms which may be responsible for the peak value in zero magnetic-field specific heat. On the basis of statistical analysis we believe that single magnetic ions, especially in the 0.024 sample case, constitute the largest part of the magnetic system. The question arises whether Mn ions in PbMnTe are really objects with a sixfold degenerate ground state, as it is usually assumed. It turns out that this is not the case. We have found[25] that the hybridization of the $3d$ electrons of manganese with the band carriers leads to a splitting of the sixfold degenerated level into, in general, three doublets. Such a splitting (without analysis of its detailed mechanism) has been already considered as the cause of the behavior of zero magnetic-field specific heat in CdMnSe.[26]

In $Pb_{1-x}Mn_xTe$ crystal the ground state of manganese is the Mn^{2+} configuration with five electrons in the $3d$ shell with orbital momentum $L=0$. Such a state is not influenced by any crystal field. However, if we allow for the hybridization we must also take into account the manganese ion in its excited Mn^{3+} and Mn^{1+} states for which $L=2$. Due to the internal spin-orbit interaction in these states we obtain an effective coupling between spin degrees of freedom and the crystal field, which leads to the ground-state splitting. The magnitude of this splitting strongly depends on the symmetry of the surroundings of the Mn ion. If the Mn ion is in a site of perfect octahedral symmetry, the sixfold degenerate level splits into a doublet and quartet. The energy distance between these two levels is of the order 10^{-4} K, which is too small to be relevant to the range of energies that correspond to our magnetic specific-heat measurements. However, such an octahedral symmetry of the manganese surroundings may be expected only in very dilute PbMnTe samples. Due to the large difference between ionic radii of Pb and Mn, the PbMnTe lattice is locally deformed and this deformation is not limited to the unit cell but spreads over larger distances. Even for the 0.024 sample the deformations originating from nearest manganese ions overlap. Because the Mn ions are placed randomly, we may expect deviations of the direction of bonds from those in the perfect lattice. According to the numerical simulations of the deformed lattice of $Pb_{1-x}Mn_xTe$ that we have performed, these deviations are of the order $1-5°$. They have a dramatic effect on the splitting of the Mn level which becomes of the order of 0.1–1 K.

We have performed a number of simulations taking into account this hybridization induced splitting. However, the calculated peak in zero magnetic-field specific heat never attained the height of the one measured in experiment. This is a consequence of Kramers' theorem which states that without an external magnetic field the levels of an odd-number electron system must be at least doubly degenerate. The $3d$ electrons of Mn which provide 5/2 spin constitute such a system and whatever the symmetry of the neighborhood was, in calculations, the ground state of the Mn ion remained doubly degenerate. Now, from thermodynamics we know that

$$\int_0^\infty dT \frac{C_H(T)}{T} = k_B N_{Mn} \ln(2S+1) - k_B \ln(g_0), \qquad (1)$$

where N_{Mn} is the number of manganese ions, $S=5/2$, $C_H(T)$ is the magnetic specific heat at temperature T, and g_0 is the degeneracy of the ground state of the system. In an external magnetic field $g_0=1$ and the second term on the right-hand side disappears.

We cannot calculate exactly the above integral using experimental data because we have no measurements below 0.5 K. However, for both samples, *assuming* a linear behavior of the specific heat at lowest temperatures, our calculations yield the Mn concentration resulting from Eq. (1) in perfect agreement with the total Mn content (0.056 in the 0.056 sample case and 0.020 for the 0.024 sample). Moreover, comparing the integrals for zero and nonzero (0.5 T) magnetic field, we obtain nearly the same results. This is particularly well seen for the 0.056 sample by looking at the experimental data. If there had been significant percentage of magnetic ions with a doubly degenerate ground state, then in zero magnetic field the degeneracy of the ground state would have been greater than 1 (in that case $g_0=2^{N_s}$, where N_s is the number of single magnetic ions with doubly degenerate ground state) and the difference between the values of the integrals for zero and nonzero magnetic field would have been much larger.

For example, the long dashed line in Fig. 5 shows the calculated magnetic specific for $x=0.024$ sample with nearest-neighbor pairs, triples, and singles split into three doublets. The energy distances between these doublets were 0.2 and 0.4 K. Again, the calculated specific heat is much smaller than the measured one.

Summarizing, we have not been able to explain the experimental results by considering the manganese spin system alone. Below we introduce a mean-field-like model in which the interaction between the manganese spin system and that of free carriers is taken into account. Models of this kind were recently applied in the studies of paramagnetic-ferromagnetic transition in semimagnetic semiconductors.[27,28]

We consider a system consisting of N_{Mn} manganese 5/2 spins and N_{el} quasifree carriers in a magnetic field applied along the z axis and described by the following model Hamiltonian:

$$H = g\mu_B B(M_z + s_z) + \frac{\alpha_0}{N_c}\mathbf{s}^2 - \frac{J_0}{N_c}\mathbf{M}\cdot\mathbf{s}. \qquad (2)$$

where $\mathbf{M}=\sum_i^{N_{Mn}}\mathbf{M}_i$ and $\mathbf{s}=\sum_j^{N_{el}}\mathbf{s}_j$ are the total spin of the manganese and electron spin system, respectively. N_c is the number of cation sites in the considered volume V of the crystal and the coefficient $\alpha_0/N_c=1/(\rho V)$ where ρ is the density of states at the Fermi level. The first term in the Hamiltonian describes the Zeeman energy of Mn spins and free carriers. The second term describes, in the lowest approximation, the energy cost of the spin polarization of the electron gas. The p-d or s-d coupling between Mn spins and free carriers is described by the last term of the Hamiltonian.

The energy levels may be characterized by the set of quantum numbers (M,s,J,J_z) as follows:

$$E(M,s,J,J_z) = -\frac{J_0}{2N_c}[J(J+1) - M(M+1) - s(s+1)]$$

$$+ g\mu_B B J_z + \frac{\alpha_0}{N_c} s(s+1), \qquad (3)$$

where $0 \leqslant s \leqslant s_{max} = N_{el}/2$, $0 \leqslant M \leqslant M_{max} = 5N_{Mn}/2$, $M-s \leqslant J \leqslant M+s$, and $-J \leqslant J_z \leqslant J$. Although the spectrum of the Hamiltonian is known, the thermodynamic quantities, in particular the magnetic specific heat, may be calculated only numerically for finite system.

In Fig. 5 we present the results of calculations for the system consisting of 2120 Mn spins and 20 electron spins, corresponding to $Pb_{1-x}Mn_xTe$ sample with $x=0.024$ and a hole concentration of 3×10^{18} cm^{-3}. The calculations were carried out for the following values of parameters: the p-d exchange integral $J_0=5\times10^3$ K and $\alpha_0=9.4\times10^4$ K.

We have checked that the behavior of the system consisting of a large number of spins described by Hamiltonian (2) remains the same if, in our model, we replace Heisenberg spins in Eq. (2) by Ising spins. In the case of Ising spins most of the calculations of thermodynamic quantities may be performed analytically and this enables us to calculate the magnetic specific heat in the thermodynamic limit, i.e., for the infinite system. It turns out that the temperature position of the peak and its height are almost the same as calculated previously for the finite system.

The model presented above shows that the exchange interaction between free carriers and localized manganese moments provides a plausible physical mechanism fully removing the degeneracy of the ground state of Mn ions and producing the magnetic specific heat comparable with experimental observations. However, its closer examination shows that quantitatively it does not work well in $Pb_{1-x}Mn_xTe$, at least in the simple form presented above. In PbMnTe, for hole concentration 3×10^{18} cm^{-3} the density of states at the Fermi level equals about 0.2×10^{17} K^{-1} cm^{-3} and the exchange integral J_0 is of the order of 200 meV. For these values of the parameters the peak in the temperature dependence of magnetic specific heat of PbMnTe is located at very low temperatures. To achieve the semiquantitative agreement between calculations and experiment (the dashed-dotted line presented in Fig. 5) we adopted

the following values of density of states: $\rho = 0.13 \times 10^{18}$ $K^{-1}\,cm^{-3}$ (about six times larger) and $J_0 = 430$ meV (about two times larger), which is rather unrealistic.

The effect of the *p-d* exchange interaction can be viewed as an action on Mn ions of effective molecular field produced by the spin polarization of conducting carriers. Our experimental data indicate that this molecular field is of the order of 0.5 T for $x = 0.024$ and 1 T for $x = 0.056$. One may expect that the cluster models may provide a good description of the experiment only in the case of external magnetic fields larger than the internal molecular field. At zero external field the cluster models should break down, as experimentally observed in PbMnTe.

We would also like to point out that in our model the molecular field is independent of Mn content. Therefore our model, without additional assumption concerning the dependence of *p-d* exchange integral on Mn content, does not explain the increase of molecular field with increase of Mn concentration.

IV. DISCUSSION AND CONCLUSIONS

The important experimental finding revealed in our measurements of the specific heat of PbMnTe concerns the unexpectedly large magnetic specific heat observed at zero external magnetic field. This result is in contrast to previous studies of magnetic specific heat of cubic II-VI SMS's with Mn and strongly suggests that all magnetic ions have the degeneracy of their ground state lifted with the energy splitting of the order of 1 K. The experimental curves for magnetic specific heat obtained at zero and small (0.5 T) external magnetic field are very similar. This suggests that certain internal magnetic field exists and that this field is responsible for the manganese ground-state splitting. However, the origin of such a field at present is not clear. The model described in the previous section does provide a mechanism for such splitting as caused by the *p-d* exchange interaction. Although the qualitative conclusions of this model are in accordance with the experiment, the model fails to provide a quantitative description of the experimental data.

On the other hand one should realize that the model is indeed very simple. It does not take into account a number of factors which may influence its predictions. First, the spatial dependence of the spin density in both systems was neglected. As it was pointed out in Ref. 28, in such an approach the spatial spin-spin correlations cannot be properly handled. In particular, there is no place in the model for the spin-wave-like magnetic excitations. Second, the model completely neglects the positional disorder in the crystal. As it was shown recently,[29] in a similar model, the positional disorder changes significantly the thermodynamic quantities. Moreover, it was assumed that the manganese ions form "ideal" spin-only $S = 5/2$ magnetic moments with a sixfold degenerate ground state. The discussion concerning the influence of the lattice deformation on the Mn spin splitting shows that this might not be a valid assumption in the subkelvin energy range. We would also like to point out the importance of the details of the energy structure of $Pb_{1-x}Mn_xTe$ in magnetic field. In particular the role of the $4s$ states of Mn, particularly for higher concentration of manganese, remains unknown. As it was explicitly shown,[30] these states play an important role in calculations of *p-d* exchange integral, even in vanishing Mn concentration limit. One should also notice the fact that the results of the optical experiments yield the *p-d* exchange integral strongly increasing with decreasing temperature.[3] This unexplained effect is not taken into account in our model.

Finally, we would like to mention a possibility to explain our experimental findings by considering the molecular field produced not by the spin polarization of conducting carriers but proportional to the magnetization of Mn ions subsystem. In the temperature range studied by us the PbMnTe crystals are paramagnetic and, consequently, the usual Weiss molecular field must be zero. In Ref. 20 the authors suggest a possibility of a spin-glass transition at subkelvin temperatures. Such a spin-glass transition could give a contribution to the specific heat even above the transition temperature. However, the essential part of our experimental data corresponds to the temperatures well above the possible spin-glass transition and our magnetic susceptibility data are perfectly regular down to 1.3 K. Therefore we did not exploit this line of interpretation in our paper.

In conclusion, we have measured the magnetic contribution to the specific heat of the semimagnetic semiconductor $Pb_{1-x}Mn_xTe$ with $x = 0.024$ and $x = 0.056$. We have analyzed the experimental data within the framework of a number of different theoretical models, applying both cluster models, developed before for semimagnetic semiconductors, as well as different models taking into account distortion of the lattice and the *p-d* exchange interaction. Our analysis clearly shows that to semiquantitatively explain the experimental data one has to develop a model which predicts about a 1-K splitting of the ground-energy state of single Mn ions in PbMnTe. We suggest that such a splitting may be caused by the *p-d* exchange coupling.

ACKNOWLEDGMENTS

We acknowledge helpful discussions with Professor J. R. Anderson. One of the authors (A.Ł.) would like to thank Professor Ł. A. Turski for very useful discussions and suggestions concerning the theoretical part of the paper. This research was supported in part by the United States Army through its European Research Office Contract N68171-00-M-5929.

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