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MAGNETIC SPECIFIC HEAT OF $(Cd_{1-x-y}Zn_yMn_x)_3As_2$

H. Bednarski[a], J. Cisowski[a], D. Schmitt[b], J. Voiron[b], J.C.Portal[c] and W. Lubczyński[a]

[a] Department of Solid State Physics, Polish Academy of Sciences Wandy 3, 41-800Zabrze, Poland
[b] Laboratoire de Magnétisme Louis Néel, CNRS, Grenoble, France
[c] Service National des Champs Intenses, CNRS, Grenoble and Département de Génie Physique, INSA, Toulouse, France

The specific heat of $(Cd_{1-x-y}Zn_yMn_x)_3As_2$ with low Mn concentration ($x \leq 0.067$) and for two Zn contents ($y = 0.14$ and $y = 0.34$) has been measured in the temperature range of 1.5–30 K. The magnetic contribution to the total specific heat has been analysed within our generalized pair approximation model which takes into account the complicated tetragonal crystal structure of $Cd_{1-x-y}Zn_yMn_x)_3As_2$. Assuming that the total Mn–Mn interaction strength is a sum of superexchange and the Bloembergen–Rowland exchange, we have obtained a very good agreement between our approach and experiment by using the analytical formulae with only two adjustable parameters, i.e. the first nearest-neighbour exchange constants for both mechanisms which appear to be strongly dependent on Zn content.

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$(Cd_{1-x-y}Zn_yMn_x)_3As_2$ (CZMA) together with $(Cd_{1-x}Mn_x)_3As_2$ (CMA) and $(Zn_{1-x}Mn_x)_3As_2$ (ZMA) belong to the family of semimagnetic semiconductors (SMSCs) based on the tetragonal II–V compounds Cd_3As_2 and Zn_3As_2. An analysis of the magnetic properties of CMA [1] and ZMA [2] (which can be treated as special cases of CZMA) has been performed within an extended version of the pair approximation, neglecting, however, the complexity of the crystal structure of both systems. The values of the nearest-neighbour (NN) Mn–Mn exchange interaction constant J_{NN} (divided by the Boltzmann constant k_B), as found in Refs. [1, 2], are equal to -100 K and -30 K, respectively, suggesting their strong dependence on Zn content. For this reason and being also interested in Mn-alloyed II–V compounds (see e.g. [3]), we have undertaken a more detailed study of the magnetic properties of CZMA, including magnetization [4] and the first specific heat data presented in this paper.

The specific heat measurements were performedt for a few samples of CZMA with different compositions, in the temperature range of 1.5–30 K. Some representative results (limited to 8 K) for $y = 0.34$ and including the sample without Mn

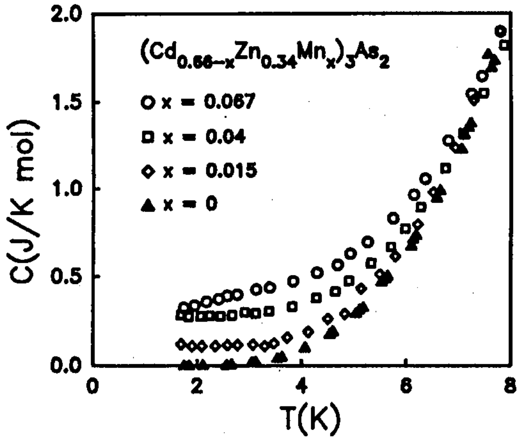

Fig. 1. Total specific heat of $(Cd_{1-x-y}Zn_yMn_x)_3As_2$ with $y = 0.34$ and for several Mn concentrations.

($x = 0$) are shown in Fig. 1. It can be seen that the presence of Mn ions strongly influences the measured specific heat which is a sum of the magnetic and lattice contributions; the role of the former contribution C_{m} can be clearly observed at low temperatures, while at higher temperatures all the data converge, indicating domination of the lattice contribution.

In order to extract C_{m} from the measured total specific heat, we have used a phenomenological interpolation procedure for the lattice simulation (PLS) [5] exploiting the fact that the atomic masses of Mn and Zn (equal to 55 and 65, respectively) do not differ appreciably. The final results for all the samples studied are presented in Fig. 2, giving a possibility to compare the data for similar Mn concentrations and for two Zn contents, i.e. $y = 0.14$ and $y = 0.34$.

While analysing the magnetic specific heat data shown in Fig. 2, we have used our approach, which may be called the generalized pair approximation (GPA) [4, 6]. The pair approximation in its usual or extended version [1, 2] may be applicable, in principle, to SMSCs with simple crystal structures in which each cation site has the same arrangement of other cations, which results in a unique NN distance between the ions. Bearing in mind the complexity of the crystal structure of II–V compounds, including also those alloyed with Mn [7, 8], we have generalized the pair approximation for an arbitrary structure. In the case of CZMA, we deal with the tetragonal structure isomorphic with phase α'' of Cd_3As_2 [9] and characterized by 3 inequivalent cation sites. These sites may be occupied at random by Mn ions, giving 4 possible NN distances. Taking these features of the crystal structure of CZMA into account and assuming that the total interaction strength $J(R)$ (where R is a distance between the Mn ions) is a sum of superexchange $J^{\mathrm{SE}}(R)$ and the Bloembergen–Rowland (BR) exchange $J^{\mathrm{BR}}(R)$ [10, 11] described by simple analytical formulae cited in Refs. [4, 6], we have obtained the theoretical curves shown in Fig. 2 with only two adjustable parameters which are the first NN con-

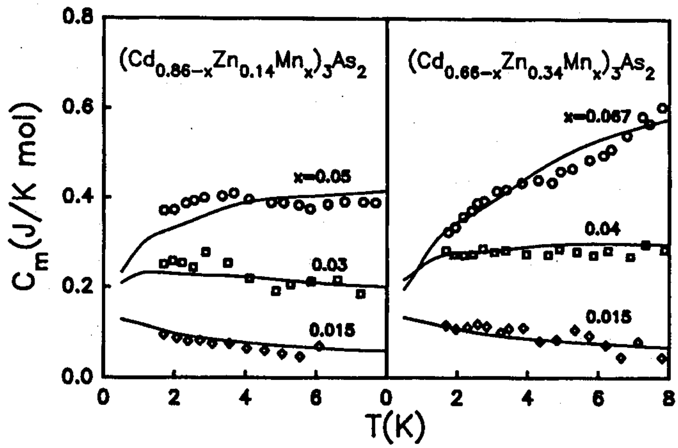

Fig. 2. Magnetic specific heat of $(Cd_{1-x-y}Zn_yMn_x)_3As_2$ with various compositions. Solid lines represent the generalized pair approximation (GPA) curves calculated with the following first NN constants (all are negative and given in K): $J_1 = J_1^{SE} + J_1^{BR} = 19K + 5.5K = 24.5K$ for $y = 0.14$ (on the left) and $J_1 = J_1^{SE} + J_1^{BR} = 25K + 7K = 32K$ for $y = 0.34$ (on the right).

stants for both mechanisms, i.e. J_1^{SE} and J_1^{BR}. Moreover, we have performed our fitting procedure in such a way that we get the experimental results for the sum $\sum_p z_p J_p = -240$ K and -304 K for $y = 0.14$ and0.34, respectively (where z_p is the number of cation sites on consecutive coordination spheres) as determined from independent susceptibility measurements performed on the same samples [12].

The final values of the exchange constants of CZMA, as listed in the caption for Fig. 2, together with the values of $J_1 = J_1^{SE} + J_1^{BR} = 12K + 4K = 16K$ and $J_1 = 53K + 11K = 64K$ obtained from our reinterpretation [4, 6] of the data for CMA [1] and ZMA [2], respectively, indicate that J_1^{SE} (giving the main contribution to the total first NN constant J_1) strongly increases with Zn content. This, in turn, can be explained, from one side, by the fact of the greater amount of Zn in CZMA, the higher degree of p–d hybridization [2, 10] and, from the other side, by a decrease in the first NN distance R_1 which changes from 3.3 Å to 3 Å when passing from CMA to ZMA.

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