Vol. 87 (1995) *ACTA PHYSICA POLONICA A* No. 1

Proceedings of the XXIII International School of Semiconducting Compounds, Jaszowiec 1994

INFLUENCE OF PRESSURE ON MAGNETIZATION OF $(Cd_{1-x-y}Zn_yMn_x)_3As_2$

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The influence of hydrostatic pressure up to 0.6 GPa on magnetization of $(Cd_{1-x-y}Zn_yMn_x)_3As_2$ with various compositions was studied at 4.2 K and in magnetic fields up to 7 T. The obtained experimental data were analysed within our generalized pair approximation model, treating the total Mn–Mn interaction strength as a sum of superexchange and the Bloembergen–Rowland exchange. As a result, we obtained satisfactory agreement between our approach and experiment by introducing a pressure dependence of p–d hybridization potential V_{pd} in the form $V_{pd} \propto d^{-4}$, where d is the Mn–As bond length which decreases with pressure.

PACS numbers: 75.30.Cr

$(Cd_{1-x-y}Zn_yMn_x)_3As_2$ (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 (SMSC's) based on the tetragonal II_3–V_2 semiconducting compounds Cd_3As_2 and Zn_3As_2. The former has an inverted band structure with the energy gap $E_g = -0.1$ eV, whereas the latter has a simple gap with $E_g = 1.1$ eV; thus, for small Zn and Mn contents, CZMA is a narrow-gap material and exhibits a linear dependence of the energy gap on composition [1].

In our previous works, we have presented the experimental studies of magnetic properties of CZMA such as high-field magnetization [2], magnetic specific heat [3] and susceptibility [4]. These quantities have been consistently interpreted within our generalized pair approximation (GPA) [5] and the values of the first nearest-neighbour (NN) Mn–Mn exchange constant (J_1) have been estimated from a fitting procedure as being equal to −16 K, −24.5 K, −32 K and −64 K for $y = 0$, 0.14, 0.34 and $1 - x$ ($y \geq 0.9$), respectively. These results suggest that the interaction strength strongly increases with Zn content and we interpret this effect as being due to a higher degree of p–d hybridization and, to a lesser extent, by a decrease in the first NN distance R_1 which changes from 3.2 to 2.9 Å, when passing from CMA to ZMA. To test our model of the d–d interaction in Mn-alloyed II_3–V_2 compounds (which is strongly correlated with the p–d interaction [6]), we have undertaken the pressure measurements of magnetization since high-pressure experiments provide a unique opportunity for studying the effect of increasing p–d hybridization in SMSC's [7].

The measurements were performed for a few samples of CZMA with $0.015 \leq x \leq 0.067$ and $0.14 \leq y \leq 0.34$. Some experimental data collected at normal pressure and at $P = 0.6$ GPa are shown in Fig. 1. It can be seen that, for all samples studied, magnetization decreases with pressure and this decrease depends on both Zn and Mn contents, as discussed at the end of this work.

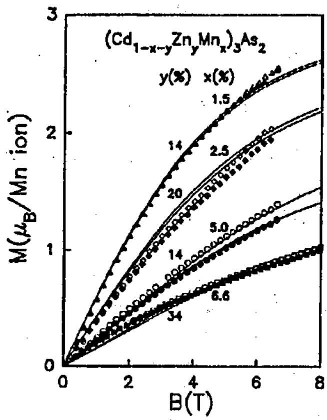

Fig. 1. Magnetization of $(Cd_{1-x-y}Zn_yMn_x)_3As_2$ with various compositions at normal pressure and at $P = 0.6$ GPa (open and closed symbols, respectively). Solid lines represent the generalized pair approximation (GPA) curves calculated with the following first NN constants at normal pressure (all are negative and given in K): $J_1 = J_1^{SE} + J_1^{BR} = 19 + 5.5 = 24.5$ for $y = 0.14$ and $J_1 = J_1^{SE} + J_1^{BR} = 25 + 6 = 31$ for $y = 0.2$ and $y = 0.34$. Influence of pressure on these constants is taken into account according to Eqs. (4)–(6).

In order to analyse the magnetization data presented in Fig. 1, we used our generalized pair approximation [5], taking into account the complexity of the crystal structure of CZMA, isomorphic with phase α'' of Cd_3As_2 [8] and characterized by 3 inequivalent cation sites. Within the GPA, magnetization M is derived from the total free energy F of the system ($M = -[\partial F/\partial B]_T$) which, in the presence of an external magnetic field B and pressure P, can be expressed as

$$F = \frac{N}{t}\sum_{j=1}^{t}\sum_{\nu_j}^{\infty} P_{\nu_j}(x)F_{\nu_j}(B, J_{\nu_j}(P)), \tag{1}$$

where $F_{\nu_j}(B, J_{\nu_j})$ is the free energy of a pair of interacting spins, $J_{\nu_j}(P) = J(P, R_{\nu_j})$ is an exchange constant between these spins separated by a distance

R_{ν_j}, $P_{\nu_j}(x)$ is the probability of finding such a pair, N is the total number of spins and t is the number of inequivalent cation sites.

Following our previous works on CZMA [2–4], we assumed that the radial dependence of the total interaction strength J for a pair of spins is a sum of superexchange J^{SE} and the Bloembergen–Rowland (BR) exchange J^{BR} [7, 9] which are now also functions of pressure and can be written as

$$J^{\mathrm{SE}} = J_1^{\mathrm{SE}}(P)\exp[-\alpha_{\mathrm{SE}}(r^2-1)], \tag{2}$$

where $r = R/R_1$ is a distance between the Mn-ions (in units of the first NN distance R_1) and $\alpha_{\mathrm{SE}} = 5.16R_1^2/a^2$, with a being the lattice constant, and

$$J^{\mathrm{BR}} = \frac{J_1^{\mathrm{BR}}(P)}{r^3}\exp[-\alpha_{\mathrm{BR}}(P)\cdot(r-1)] \tag{3}$$

with $\alpha_{\mathrm{BR}}(P) = R_1(P)(2m_{\mathrm{e}}E_{\mathrm{g}})^{1/2}/h$, where m_{e} and E_{g} are the electron effective mass and the energy gap, respectively. It is interesting to note that the quantities r and α_{SE} are pressure independent, being both the ratios of any distance d which changes uniformly under the effect of hydrostatic pressure as

$$d = d_0\left(1+\frac{1}{3}KP\right), \tag{4}$$

where K is the compressibility taken for CZMA as that deduced for Cd_3As_2, i.e. $K = -0.022$ GPa^{-1} [10]. Thus, the total interaction strength $J = J^{\mathrm{SE}} + J^{\mathrm{BR}}$ contains only two unknown parameters, i.e. the first NN constants for both mechanisms J_1^{SE} and J_1^{BR} which can be determined by a fitting procedure [2–4]. However, as follows from theoretical works [6, 11], both J_1^{SE} and J_1^{BR} are proportional to V_{pd}^4, where V_{pd} is the p–d hybridization potential. This potential, in turn, is proportional to d^{-4} [12, 13], where d is now the Mn–As bond length which changes with pressure according to Eq. (4). Thus, combining all these relationships together with Eqs. (2)–(4), one can write the pressure dependence of the first NN exchange constants for superexchange and the BR exchange as

$$J_1^{\mathrm{SE}}(P) = J_1^{\mathrm{SE}}(0)\left(\frac{d}{d_0}\right)^{-16} \tag{5}$$

and

$$J_1^{\mathrm{BR}}(P) = J_1^{\mathrm{BR}}(0)\left(\frac{d}{d_0}\right)^{-19}. \tag{6}$$

The final results of our calculations of magnetization as a function of magnetic field and pressure within the GPA are shown in Fig. 1 as solid lines, giving a satisfactory description of the experimental data. The values of the exchange constants at normal pressure, i.e. $J_1^{\mathrm{SE}}(0)$ and $J_1^{\mathrm{BR}}(0)$, used in these calculations are the same as those found previously for a given Zn content [2–4] and are listed in the caption for Fig. 1. At $P = 0.6$ GPa, these constants increase to $J_1 = J_1^{\mathrm{SE}} + J_1^{\mathrm{BR}} = 20.4\ \mathrm{K} + 6\ \mathrm{K} = 26.4\ \mathrm{K}$ for $y = 0.14$ and $J_1 = 26.8\ \mathrm{K} + 6.5\ \mathrm{K} = 33.3\ \mathrm{K}$ for $y = 0.34$.

Basing on the obtained results, one can roughly explain the effect of pressure on magnetization of samples of CZMA with various compositions as follows.

The statistics of Mn ions for a given crystal structure is pressure independent and determined only by their concentration x (see Eq. (1)). Therefore, since the superexchange contribution to the total exchange significantly increases with Zn content (y) and is less sensitive to pressure (see Eqs. (5)–(6)), the resulting behaviour of magnetization for higher y is also less sensitive to pressure as can be really observed in Fig. 1, when comparing samples with similar values of x (5 and 6.6%), but with different values of y (14 and 34%). In the opposite case, i.e. for a fixed y and when changing x, the statistics comes into play which results in increasing number of interacting pairs of Mn ions as x increases and this, in turn, causes magnetization of a sample with higher x to be more sensitive to pressure than for lower x, as can be seen in Fig. 1 for two samples with the same value of y (14%), but with different values of x (1.5 and 5%).

Finally, it is noteworthy that such a significant increase (more than 7%) of the exchange constants follows mainly from a small corresponding decrease (of about 0.44% at 0.6 GPa) of the interionic distance d, showing usefulness of the pressure methods for studying the magnetic interactions in SMSC's.

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