Cryobaric exciton magnetophotoluminescence in $Cd_{1-x}Mn_xSe$

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Abstract The mechanism of the distant-neighbor exchange interactions in a diluted magnetic semiconductor CdMnSe is studied on the basis of the pressure dependence of the energy shift of excitons under magnetic field.

1 Introduction

In diluted magnetic semiconductors of II-VI and III-V compounds the magnetic ions are scattered throughout the cation sublattice of the network of sp^3 covalent bonds. The majority of the magnetic ions are isolated and the rest form small clusters, i.e., pairs, triads, quartets and so on. The spin interactions among the magnetic ions depend strongly on the energy gap and carriers of the host semiconductor. For pairs in $Cd_{1-x}Mn_xSe$ the cryobaric study[1] of the exciton magnetophotoluminescence has shown that the interaction between the two Mn^{2+} spins constructing a pair can be described in terms of the kinetic exchange theory based on the three-level model of Larson et al.[2], the model being comprised of the upper and lower Hubbard states of localized *d* electrons and the valence band of extended anion *p* orbitals. At present, however, the mechanism of spin interactions among the small clusters themselves is to be studied yet.

In the present study we examine the properties of the *n*th-neighbor exchange energies J_ns of $n \geq 2$ in $Cd_{1-x}Mn_xSe$ of x=0.001, 0.05 and 0.10 by the cryobaric magnetophotoluminescence spectroscopy.

2 Experiment and results

We measure the pressure dependence of the near-gap magnetophotoluminescence spectrum at liquid helium temperatures using an optical system[3] consisting of a diamond anvil cell of clamp type and fiber optics. A hydrostatic environment is obtained using the condensed Ar as the pressure-transmitting medium. The steady magnetic field up to 23 T is generated with a hybrid magnet.

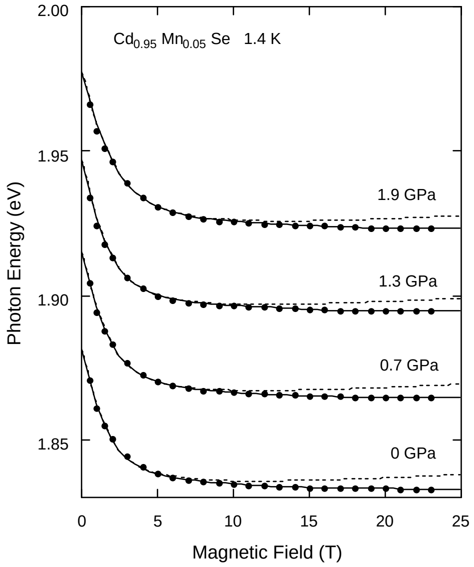

Fig. 1 Magnetic-field-induced energy shift of excitons in $Cd_{0.95}Mn_{0.05}Se$ at 1.4 K at several pressures. The magnetic field is applied parallel to the *c*-axis. The solid lines are the theoretical curves. The dotted lines are the curves calculated with the Mn^{2+}-pair contributions subtracted.

Figure 1 shows the magnetic-field-induced shift of the photoluminescence energy of excitons observed for the crystal of x=0.05 at 1.4 K under several pressures. The shift arises mainly from coupling of spins between excitons and Mn^{2+} ions[4]. The initial rapid shift reflects the magnetization of singles and triads of Mn^{2+} ions. The slope scales with the lattice temperature T and an effective temperature T_0 of Mn^{2+} spins as $(T+T_0)^{-1}$. If the magnetic field exceeds 10 T the influence of the staircasewise magnetization of pairs manifests itself as an additional shift. From the pair component we obtain $J_1/k = -7.4\pm0.4$ K at 1 atm and the pressure coefficient to be $d\ln|J_1|/dP = 0.25\pm0.05\ \mathrm{GPa}^{-1}$.

The data shown in Fig. 1 give T_0=2.0±0.2 K at 1 atm. Furthermore, T_0 remains positive under high pressures. The positive value of T_0 originates from antiferromagnetic internal field due to distant-neighbor spins. Consequently, T_0 is elevated prominently as the Mn content x increases, as shown in Fig. 2. We find that T_0 is elevated also by pressure as shown in Fig. 3.

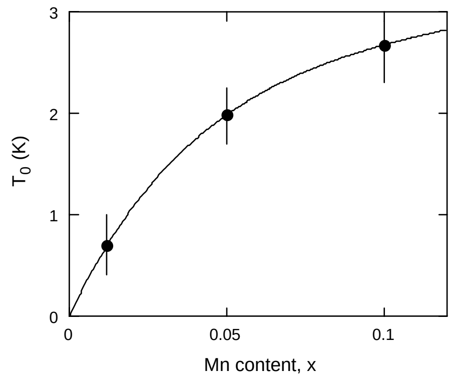

Fig. 2 The x dependence of T_0 in $Cd_{1-x}Mn_xSe$ at 1 atm. The solid line is the theoretical curve.

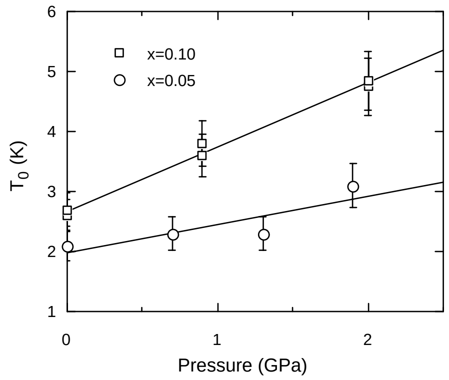

Fig. 3 Pressure dependence of T_0 in $Cd_{1-x}Mn_xSe$.

3 Discussion

In the mean-field approximation T_0 is expressed as[5]

$$kT_0 = -4p_1{}^*S(S+1)J^*, \qquad (1)$$

with an effective internal exchange constant J^* of

$$J^* = J_2 + \frac{10}{3}J_3 + 2J_4. \qquad (2)$$

The quantity $p_1{}^*$ in Eq.(1) is the probability that a Mn^{2+} ion behaves as singles. With the formula for $p_1{}^*$ given by Shapira[6], the experimental data shown in Fig.2 yield $J^*/k = -1.9\pm0.4$ K at 1 atm. From the pressure dependence of T_0 we obtain $d\ln|J^*|/dP = 0.24\pm0.1$ and 0.4 ± 0.1 GPa^{-1} for x=0.05 and 0.10, respectively.

The experimental values of $J_1/k = -7.4$ K and $J^*/k = -1.9$ K at 1 atm permit us to make a test of the validity of various proposals on the variation of J_n with n. For example, Larson's formula[2], $J_n = J_0\exp(-2.45r_n{}^2)$, claims $J^*/k = -0.85$ K, where r_n is the nth-neighbor distance normalized by the nearest-neighbor distance. Twardowski's[7] and Rusin's[8] power laws, $r_n{}^{-6.8}$ and $r_n{}^{-8.5}$, give J^*/k = -1.47 and -0.68 K, respectively, whereas Shen's independent-exchange-path model[9] gives J^*/k = -4.1 K if Shen's γ-parameter of 0.044 is adopted. It appears that Twardowski's power law is in accord with the case of diluted CdMnSe well.

We note from our experimental results that the pressure coefficient of J^* agrees with that of J_1 within the experimental errors. Although Larson's three-level model underestimates J^*, the above-mentioned exponential law predicts that the relative magnitudes of intersite spin interactions are independent of the crystal volume. In contrast the chemical bond picture such as the independent-exchange-path model presumes multiple super-exchanges along the chemical bonds connecting two spins, implying that the pressure coefficient of J^* is larger than twice that of J_1. In this sense our experimental results are in favor of Larson's picture of covalent spin interactions.

Acknowledgements

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