EXCITON MAGNETOPHOTOLUMINESCENCE STUDY ON THE Mn-Mn PAIR INTERACTION IN $Cd_{0.95}Mn_{0.05}Se$ UNDER HIGH PRESSURES

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ABSTRACT

The pressure dependence of the exciton magnetophotoluminescence is studied in $Cd_{0.95}Mn_{0.05}Se$ with attention focused on the pressure effects on the stepwise magnetization of the nearest-neighbor Mn-Mn pairs. The Mn-Mn superexchange interaction, as well as the exciton-Mn exchange interaction, is found to be strengthened by pressure. The pressure coefficients of the exchange constants indicate that the effective Coulomb energy U of Mn ions and the charge-transfer energy Δ are reduced by pressure.

1. Introduction

It has emerged from recent magnetophotoluminescence studies of II-VI diluted magnetic semiconductors that the exchange interaction between an exciton and transition-metal (*TM*) ions is strengthened by hydrostatic pressure. [1,2] Although a large part of the observed effect can be explained in terms of the bond-length dependence of the *p-d* transfer integral V_{pd}, it is suggested that the effective Coulomb energy U of d electrons of *TM* ions and/or the charge-transfer energy Δ between the upper Hubbard state of *TM* ions and the topmost edge of the valence band are also affected by pressure. [1] In order to obtain further information on this problem, we study the pressure effects on the magnetization steps[3] due to the Mn-Mn pairs in $Cd_{0.95}Mn_{0.05}Se$ by the exciton magnetophotoluminescence method.

2. Experiment

Pressure is generated with a diamond anvil cell. A hydrostatic environment is obtained by using condensed argon as the pressure-transmitting medium.The maximum pressure is limited to 2 GPa in order to sustain the wurtzite structure.The diamond anvil cell is immersed in superfluid He at 1.4 K. The static magnetic field of up to 25 T is generated with a hybrid magnet, and is applied parallel to the c-axis of the sample. The excitation and detection of the photoluminescence are performed with fiber-optic apparatus. [4]

3. Results and Discussion

Figure 1 shows the magnetic-field dependence of the photoluminescence spectrum due to the A-exciton at 0 and 1.9 GPa. In Fig.2 is plotted the field-induced energy shift δE_A of the free exciton line (F) observed under several pressures. At any pressure the energy shift is almost leveled in the field region above 6 T, since the thermal average $<S_s>$ of the spins of isolated Mn ions is saturated under high magnetic fields. We see that pressure enhances δE_A. This is due to the enhancement of the exciton-Mn exchange interaction[1,2] that is dominated by the hole-Mn kinetic spin interaction,[5] *i.e.*, the hybridization term of the *p-d* exchange interaction. In addition, the weak stepwise magnetization of the antiferromagnetically coupled Mn ions, which are substituted for the nearest-neighbor cations, can also be identified in the field dependence of δE_A above 10 T. The stepwise anomalies of magnetization occur successively at magnetic fields $H_n (n=1, 2, ..5)$ at which the total spin S_p of the ground state of the paired Mn ions increases from $n-1$ to n as a result of the energy-level crossing. The derivative $-dE_A/dH$ is plotted as a function of the external field H in the inset of Fig.2. It is clearly seen that H_1 shifts toward higher magnetic field with increasing pressure. H_n is related to the exchange coupling $-2J_{NN}\mathbf{S}_i\cdot\mathbf{S}_j$ of Mn spins by[3] $H_n=2n\,|J_{NN}|(g\mu_B)^{-1}+H_d$, where g and μ_B are the g-parameter of a Mn ion and the Bohr magneton, respectively, and $H_d \approx 1.5$ T is the correction due to the distant-neighbor Mn-Mn interactions. The experimental results shown in Fig.2 indicate that $|J_{NN}|$ is significantly enlarged by pressure.

When pairs of *TM* ions coexist with isolated ions, the mean spin $<S>$ is given by $<S>=f_1<S_s>+(f_2/2)<S_p>$ with the probabilities f_1 and f_2 that a *TM* ion is isolated and paired with another *TM*

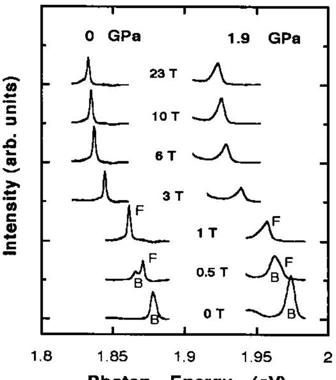

Figure. 1. Photoluminescence spectrum due to the A-exciton in $Cd_{0.95}Mn_{0.05}Se$ under various magnetic fields at 0 and 1.9 GPa. The features F and B are due to free and bound excitons, respectively.

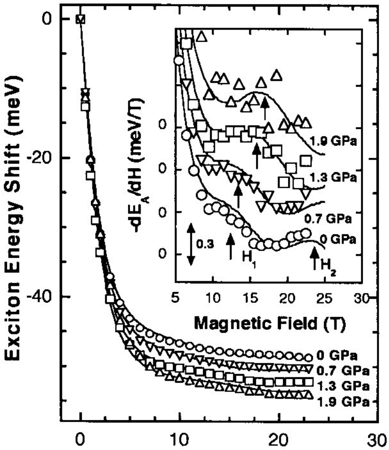

Figure 2. Field-induced energy shift of the free exciton line in $Cd_{0.95}Mn_{0.05}Se$ under 0 (○), 0.7 (▽), 1.3 (□), and 1.9 (△) GPa. The inset shows the derivative of the exciton energy with respect to the external magnetic field. The solid lines are the theoretical curves. Arrows show the field positions H_1 and H_2.

ion, respectively. For Mn^{2+} ions, $<S_s>$ is given by a modified Brillouin function for the spin of $S = 5/2$, while $<S_p>$ has been formulated in Ref.[3] as a function of H and temperature. In the case of $H \parallel c$, δE_A is given to a good approximation by

$$\delta E_A = -\frac{1}{2} |N_0\beta_{hyb}| x<S> + E_{Z,d} , \tag{1}$$

where $N_0\beta_{hyb}$ is the exchange constant of the p-d hybridization term and $E_{Z,d}$ represents the linear Zeeman and diamagnetic shifts of the A-exciton due to the external magnetic field. In Fig. 2 the theoretical curves of δE_A are compared with experimental results. The calculations are performed by taking $N_0\beta_{hyb}$ and J_{NN} as the adjustable parameters. The calculated curves agree well with the experimental results. Figure 3 shows the relative pressure dependence of $|N_0\beta_{hyb}|$ and $|J_{NN}|$ extracted from this analysis. The pressure coefficients are found to be 7.0±0.1 %/GPa and 20±2 %/GPa for $|N_0\beta_{hyb}|$ and $|J_{NN}|$, respectively. According to recent theoretical investigations,[6,7] these kinetic-exchange constants are given well by

$$N_0\beta_{hyb} = -\frac{16}{S} V_{pd}^2 \left(\frac{1}{\Delta} + \frac{1}{U-\Delta}\right), \tag{2}$$

and

$$J_{NN} = -\frac{1}{2S^2} \frac{V_{pd}^4}{\Delta^2} \left(\frac{1}{U} + \frac{1}{\Delta}\right) j. \tag{3}$$

The coefficient j in Eq.(3) is a dimensionless constant. At 1 atm we have $N_0\beta_{hyb}=-1.37$ eV[Ref. 8], $J_{NN} = -6.4\times10^{-4}$ eV, U=7.6 eV[Ref. 6], and Δ=4.2 eV[Ref. 6]. Thus Eqs.(2) and (3) yield j=2.35.

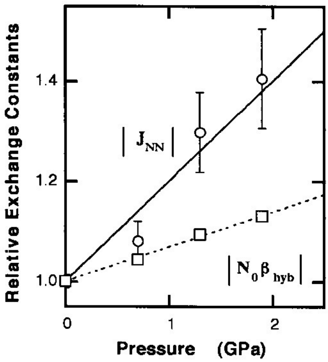

Figure 3. Pressure dependencies of J_{NN} and $N_0\beta_{hyb}$. The solid and dotted lines show the pressure coefficients of 20% /GPa and 7% /GPa, respectively.

Harrison's scaling theory[9] predicts that the transfer integral V_{pd} depends on the Mn-Se bond length l as $l^{-7/2}$. Therefore, the linear compressibility $\kappa = 0.62$ %/GPa of the lattice gives $d\ln V_{pd}^2/dP \approx 4.3$ %/GPa and $d\ln V_{pd}^4/dP \approx 8.7$ %/GPa. The discrepancies by a factor of about 2 seen in comparison with the experimental values, $d\ln|N_0\beta_{hyb}|/dP \approx 7\%$/GPa and $d\ln|J_{NN}|/dP \approx 20\%$/GPa, support the notion that U and/or Δ vary with pressure. Figure 4 shows the pressure dependencies of U and Δ calculated from Eqs.(2) and (3) with experimental values of $N_0\beta_{hyb}$ and J_{NN}, and V_{pd}^2 and V_{pd}^4 scaled from Harrison's formula. Both U and Δ turn out to be reduced by pressure at rates of −0.18 and −0.12 eV/GPa, respectively. The magnitude of the relative reduction of U, $|d\ln U/dP| \approx 2.4\%$ /GPa, is as great as the magnitude of the relative increase of V_{pd}, $d\ln|V_{pd}|/dP \approx 2.2\%$/GPa. The observed reduction of U is qualitatively explained by the enhancement of the screening effect, due to valence electrons, resulting from an increase in the *p-d* hybridization. The strong sensitivity of the exchange interactions to the change in Δ leads us to envisage that the giant magneto-optical properties of a diluted magnetic semiconductor will be affected by quantum lattice structures, since the valence band is altered by the quantum confinement of electrons.

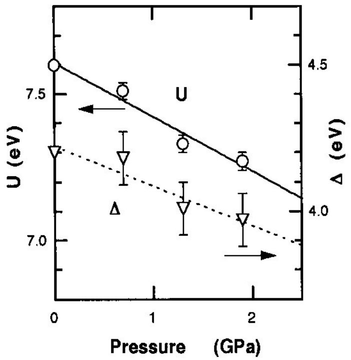

Figure 4. Pressure dependencies of U and Δ. The solid and dashed lines are the liner fits to the experimental data of U and Δ, respectively.

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References

1. Y. Matsuda, N. Kuroda, I. Mogi, G. Kido, and Y. Nishina, *Physica* B **201**, 411 (1994) ; Y. Matsuda and N. Kuroda, *Phys. Rev.* B **53**, 4471 (1996).
2. R. Meyer, M. Dahl, G. Schaack, A. Waag, and R. Boehler, *Solid State Commun.* **96**, 271 (1995).
3. S. Foner, Y. Shapira, D. Heiman, P. Becla, R. Kershaw, K Dwight, and A. Wold, *Phys. Rev.* B **39**, 11793 (1989); see also the references therein.
4. Y. Matsuda, N. Kuroda, and Y. Nishina, *Rev. Sci. Instrum.* **63**, 5764 (1992).
5. A. K. Bhattacharjee, G, Fishman, and B. Coqblin, *Physica* **117B** & **118B**, 449 (1983).
6. B.E. Larson, K.C. Hass, H. Ehrenreich, and A. E. Carlsson, *Phys. Rev.* B **37**, 4137 (1988).
7. J. Zaanen, G.A. Sawatzky, and J. W. Allen, *Phys. Rev. Lett.* **55**, 418 (1985).
8. R. L. Aggarwal, S. N. Jasperson, J. Stankiewicz, Y. Shapira, S. Foner, B. Khazai, and A. Wold, *Phys. Rev.* B **28**, 6907 (1983).
9. W. A. Harrison, *Elecronic Structure and the Properties of Solids* (Dover, New York, 1989), p.451.