Optical determination of the antiferromagnetic exchange constant between nearest-neighbor Mn^{2+} ions in $Cd_{0.95}Mn_{0.05}Te$

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Optical splittings of the band-edge exciton in $Cd_{0.95}Mn_{0.05}Te$ have been measured at 1.45 K with dc magnetic fields B up to 20 T, using magnetoreflectance in the Faraday configuration. The splitting $\Delta E_{3/2}$ between the highest- and lowest-energy components, involving $\pm\frac{3}{2}$ hole states, follows a modified Brillouin function at low values of B. At higher values of B, $\Delta E_{3/2}$ exhibits temperature-broadened, steplike increases due to the magnetic-field-induced alignment of the antiferromagnetically coupled nearest-neighbor (NN) Mn^{2+}-ion pairs. The first of the expected five steps was observed at $B_1=11.5\pm0.5$ T, which yields a value $J_{NN}=-7.7\pm0.3$ K for the Mn^{2+} nearest-neighbor antiferromagnetic exchange constant. The presence of a second step is indicated at $B_2=19.5\pm1.0$ T, somewhat below the predicted value of $2B_1$.

I. INTRODUCTION

In this paper we report the first direct measurement of the antiferromagnetic exchange constant J_{NN} between the nearest-neighbor (NN) Mn^{2+} ions in $Cd_{0.95}Mn_{0.05}Te$, using spin splittings of the band-edge free excitons in magnetoreflectance[1] at 1.45 K. We previously reported similar measurements for $Cd_{0.95}Mn_{0.05}Se$.[2]

Mn^{2+} ions in II-VI compound semimagnetic semiconductors (SMS) such as $Cd_{1-x}Mn_xTe$, $Cd_{1-x}Mn_xSe$, etc., have spin angular momentum $S=\frac{5}{2}$ which arises from the half-filled $3d$ shell.[3] These Mn^{2+} spins interact with one another through an antiferromagnetic (AF) short-range exchange interaction which is presumed to decrease rapidly with increasing distance between the Mn^{2+} ions, leading to unique magnetization (M) behavior of semimagnetic semiconductors as a function of the applied magnetic field B. As shown recently,[2,4] the NN Mn^{2+}-Mn^{2+} interaction leads to a series of five steps in M versus B above 10 T, providing a direct determination of J_{NN}. The Mn^{2+}-Mn^{2+} interaction can also be probed optically because there is a strong exchange interaction between the Mn^{2+} spins and those of the band electrons, with the free-exciton splittings being proportional to M.[1,5]

The observed free-exciton splittings include a small contribution from the direct effect of applied magnetic field on the electron and hole comprising the exciton. The diamagnetic energy of the exciton in $Cd_{0.95}Mn_{0.05}Te$, also due to the direct effect of B, is found to be comparable with that in CdTe, and in both cases are found to fit Larsen's hydrogenic model[6] for donors in a magnetic field.

II. EXPERIMENTAL

$Cd_{0.95}Mn_{0.05}Te$ and CdTe samples used in this work were single crystals grown by the Bridgman method, and were cleaved along a (110) plane. Magnetoreflectance measurements were made on the (110)-cleaved surface in the Faraday configuration with samples immersed in pumped liquid helium at 1.45 K. The optical cryostat had quartz windows tilted at an angle of 5° with respect to the sample reflecting surface in order to prevent light reflected by the windows from reaching the photodetector. A Bitter solenoid with 5.4-cm bore provided a dc magnetic field B along the [110] crystal axis, up to 20 T in the case of $Cd_{0.95}Mn_{0.05}Te$, and up to 15 T in the case of CdTe. The optical spectrometer used in this work consisted of a 0.267-m single-pass monochromator (Perkin-Elmer model 99G) equipped with a blazed reflection grating (Bausch&Lomb, $\lambda_B=0.75$ μm with 1200 grooves/mm), tungsten ribbon filament lamp (General Electric type T8 1/2), a long-wave pass filter (Corning Glass no. CS3-73), a circular sheet polarizer (Polaroid model HNCP with design wavelength of 0.56 μm), 510-Hz light-beam chopper, silicon photodiode detector (EG&G model SGD-444), and a lock-in detector (PARC model 5101).

III. THEORETICAL BACKGROUND

The crystal structure of $Cd_{0.95}Mn_{0.05}Te$ solid solution is zinc blende, the same as that of CdTe. The band-edge free excitons are formed by the excitation of electrons from the p-like Γ_8 valence band to the s-like Γ_6 conduction band. We need to consider the effect of an external magnetic field B on the $1s$ ground state of the exciton which, at $B=0$, is eightfold degenerate due to the fourfold degeneracy of the Γ_8 hole and twofold degeneracy of the Γ_6 electron. The magnetic field has two important effects on the $1s$ exciton state: (a) removal of the eightfold degeneracy of the exciton by the combined effect of the g factor splitting of the conduction- and valence-band edges (as in CdTe) and the exchange interaction of the Mn^{2+} ions with electrons and holes, and (b) an increase in the exciton energy, arising from the diamagnetic effect. Typically in SMS crystals, including $Cd_{0.95}Mn_{0.05}Te$, the exchange interaction with the magnetic ions dominates the effects due to (i) the e-h exchange interaction and (ii) terms with cubic anisotropy.[7-9] Therefore, we neglect these small effects and use the rather simple spherical approximation in considering the exciton Hamiltonian.

The electron-Mn^{2+} and hole-Mn^{2+} exchange contribution to the Hamiltonian can be written as[3,10]

$$H_{\text{exch}}=-\sum_i [J_e(\mathbf{r}_e-\mathbf{R}_i)\mathbf{s}_e+J_h(\mathbf{r}_h-\mathbf{R}_i)\mathbf{s}_h]\cdot\mathbf{S}_i \ , \quad (1)$$

where J_e (J_h) is the coupling constant for the exchange interaction between the electron (hole) of spin $\mathbf{s}_e$ ($\mathbf{s}_h$) located at $\mathbf{r}_e$ ($\mathbf{r}_h$) and Mn^{2+} ions of spin $\mathbf{S}_i$ located at $\mathbf{R}_i$.

Because the electron and hole wave functions are very extended, they "see" a large number of magnetic ions at any given time. Consequently, the exchange energy of the exciton may be written in the form

$$E_{\text{exch}}=-x\langle S_z\rangle(N_0\alpha M_e+\tfrac{1}{3}N_0\beta M_h) \ , \quad (2)$$

where x is the molar fraction of Mn^{2+} ions, $\langle S_z\rangle$ is the average value of the z component of the Mn^{2+} ion spin, N_0 is the number of unit cells per unit volume, $\alpha=\langle\psi_e|J_e|\psi_e\rangle$, $\beta=\langle X|J_h|X\rangle$, and M_e (M_h) is the z component of the electron (hole) angular momentum. In the above, ψ_e denotes the Γ_6 conduction-band-edge state, and X denotes the p-like hole state.

In order to obtain an expression for $\langle S_z\rangle$, we recall that there is an antiferromagnetic interaction among Mn^{2+} ions. The strength of this AF interaction is largest (~ 10 K) between nearest neighbors and falls off rapidly for more distant neighbors. Thus the value of $\langle S_z\rangle$ at low temperatures will depend upon the concentration and distribution of Mn^{2+} ions, in addition to the strength of the applied magnetic field. The probability P_s, P_p, P_t^0, and P_t^c for the presence of single (isolated) Mn^{2+} ions, Mn^{2+}-ion pairs, open triangle triples, and closed triangle triples can be calculated[4,11,12] on the basis of a random distribution of Mn^{2+} ions. For $Cd_{0.95}Mn_{0.05}Te$, one obtains $P_s=0.54$, $P_p=0.24$, $P_t^0=0.09$, and $P_t^c=0.02$. This accounts for 89% of the total Mn^{2+} ions, with the remainder presumably being in larger clusters. The contribution of the isolated Mn^{2+} ions and Mn^{2+}-ion clusters excluding pairs can be written as[2,4]

$$\langle S_z\rangle_{\mathscr{B}}=-\frac{5}{2}\frac{\bar{x}}{x}\mathscr{B}_{5/2}\left[\frac{5\mu_B B}{k(T+T_0)}\right] , \quad (3)$$

where

$$\frac{\bar{x}}{x}=(P_s+\tfrac{1}{3}P_t^0+\tfrac{1}{15}P_t^c)+\tfrac{1}{5}(1-P_s-P_p-P_t^0-P_t^c) \ .$$

$\bar{x}$ represents the effective molar concentration of isolated Mn^{2+} ions, $\mathscr{B}_{5/2}$ is the Brillouin function of index $\frac{5}{2}$, and T_0 represents the phenomenological AF interaction of the relatively isolated Mn^{2+} ions with distant neighbors.

The Hamiltonian for the Mn^{2+} pair in a magnetic field is given by

$$H_p=-2J_{\text{NN}}\mathbf{S}_1\cdot\mathbf{S}_2+g_{\text{Mn}}\mu_B\mathbf{S}_p\cdot\mathbf{B} \ , \quad (4)$$

where

$$\mathbf{S}_p=\mathbf{S}_1+\mathbf{S}_2 \ .$$

J_{NN} is the NN exchange constant between Mn^{2+} ions, S_1 and S_2 are the spins of the two members of the pair, and g_{Mn} is the g factor of the Mn^{2+} ion. The energy levels of the pair for the Hamiltonian of Eq. (4) are

$$E_p=-J_{\text{NN}}[S_p(S_p+1)-\tfrac{35}{2}]+g_{\text{Mn}}\mu_B M_p B \ , \quad (5)$$

where $S_p=0,1,\ldots,5$, and

$$M_p=S_p, S_p-1, \ldots, -S_p \ .$$

It follows from Eq. (5) that the ground state of the pair corresponds to $S_p=0$ for $J_{\text{NN}}<0$ and $B<B_1$, where

$$B_1=-2J_{\text{NN}}/g_{\text{Mn}}\mu_B \ . \quad (6)$$

At $B=B_1$, the $M_p=-1$ component of the $S_p=1$ state crosses the $S_p=0$ state, as schematically illustrated in Fig. 1. Therefore, we would expect a sharp step decrease in $\langle S_z\rangle$ equal to

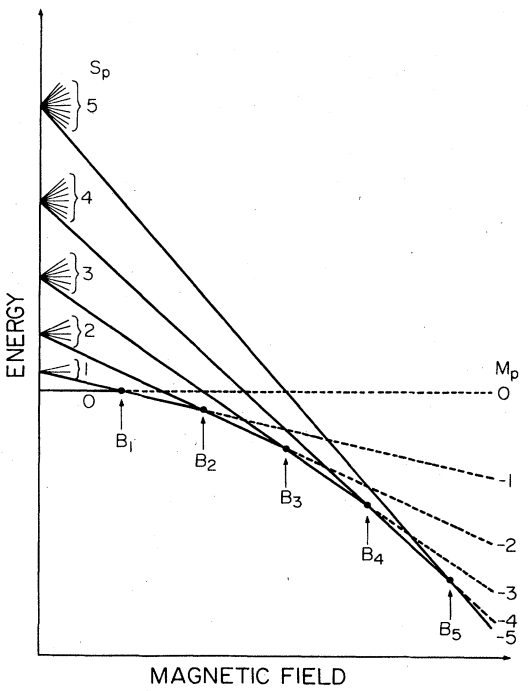

FIG. 1. Schematic of the energy levels for the antiferromagnetically coupled nearest-neighbor Mn^{2+}-ion pairs in semimagnetic semiconductors such as $Cd_{1-x}Mn_xTe$, $Cd_{1-x}Mn_xSe$, etc., in an external magnetic field.

$$\langle S_z \rangle = -P_p/2 \tag{7}$$

at $B=B_1$, assuming that $T=0$ and that the broadening due to interactions with distant neighboring ions is negligible. At $B=2B_1$, the $M_p=-2$ component of the $S_p=2$ state would cross the $M_p=-1$ component of the $S_p=1$ state, resulting in a second step decrease in $\langle S_z \rangle$ of the same size as the first. Continuing this argument, we expect a total of five steps at

$$B_n = nB_1 = -2nJ_{\mathrm{NN}}/g_{\mathrm{Mn}}\mu_B \ , \tag{8}$$

where $n=1,2,\ldots,5$. At $B=5B_1$ the pair would become completely aligned (antiparallel) with the applied magnetic field. For $T\neq 0$, the total contribution of the pairs to $\langle S_z \rangle$ can be written in the form[2]

$$\langle S_z \rangle_p = -\tfrac{1}{2}P_p \sum_n \left[1+\exp\left[\frac{g_{\mathrm{Mn}}\mu_B}{kT}(B_n - B) \right] \right]^{-1} , \tag{9}$$

where $n=0,1,\ldots,5$. In writing Eq. (9), we have assumed that only two levels near the crossing point are populated and that the relative population of these two levels is determined by the Boltzmann factor. Combining Eqs. (3) and (9), we obtain

$$\langle S_z \rangle = -\frac{5}{2}\frac{\bar{x}}{x}\mathscr{B}_{5/2}\left[\frac{5\mu_B B}{k(T+T_0)} \right] - \tfrac{1}{2}P_p \sum_n \left[1+\exp\left[\frac{g_{\mathrm{Mn}}\mu_B B}{kT}(B_n - B) \right] \right]^{-1} . \tag{10}$$

Steps are also expected for triples.[4] The first step due to open triples should occur at $B=\frac{7}{2}B_1$, where B_1 is the field, given by Eq. (6), for the first step due to a pair. The first step due to closed triples should occur at $B=\frac{3}{2}B_1$. However, we have ignored the contribution of these steps from triples because of their low concentration in $Cd_{0.95}Mn_{0.05}Te$.

The "conventional" Zeeman spin contribution (as in CdTe) to the exciton Hamiltonian is

$$H_Z = \mu_B \mathbf{B}\cdot(g_e \mathbf{s}_e - 2\tilde{\kappa}\mathbf{s}_h) \ , \tag{11}$$

where g_e is the g factor of the Γ_6 electron, and $\tilde{\kappa}$ is related[8] to the Luttinger parameter κ^L for the Γ_8 hole. The corresponding conventional Zeeman energy of the exciton is given by

$$E_Z = \mu_B B(g_e M_e - 2\tilde{\kappa}M_h) \ . \tag{12}$$

The diamagnetic contribution to the exciton Hamiltonian may be written as

$$H_d = \rho^2\gamma^2/4 \ , \tag{13}$$

where $\rho=(x^2+y^2)^{1/2}$ is the component transverse to the magnetic field of the relative position $|\mathbf{r}_e-\mathbf{r}_h|$ of the electron and hole in the exciton, and γ is a dimensionless magnetic field parameter defined as

$$\gamma = \hbar\omega_c/2\mathscr{R} = \hbar eB/2\mu c\mathscr{R} \ . \tag{14}$$

In Eq. (14) μ is the reduced mass for the electron and hole, and $\mathscr{R}$ is the effective Rydberg constant

$$\mathscr{R} = \mu e^4/2\epsilon^2\hbar^2 \ , \tag{15}$$

where ϵ is the static dielectric constant. The diamagnetic energy $E_d=\langle H_d \rangle$ has been calculated by Larsen,[6] among others, for the ground state of the hydrogen atom as a function of γ. In the following section we fit our experimental results for E_d with the variational calculations of Larsen.[6]

Selection rules for optical transitions to exciton states are $M=M_e+M_h=+1$ for σ_+ polarization, $M=-1$ for σ_- polarization, and $M=0$ for π polarization. According to these selection rules, a total of four transitions are allowed in the Faraday configuration (used in this work), corresponding to (a) $M_e=-\frac{1}{2}$, $M_h=\frac{3}{2}$, (b) $M_e=\frac{1}{2}$, $M_h=\frac{1}{2}$, (c) $M_e=-\frac{1}{2}$, $M_h=-\frac{1}{2}$, and (d) $M_e=\frac{1}{2}$, $M_h=-\frac{3}{2}$. The transitions labeled a and b occur in the σ_+ polarization, and those labeled c and d occur in the σ_- polarization.

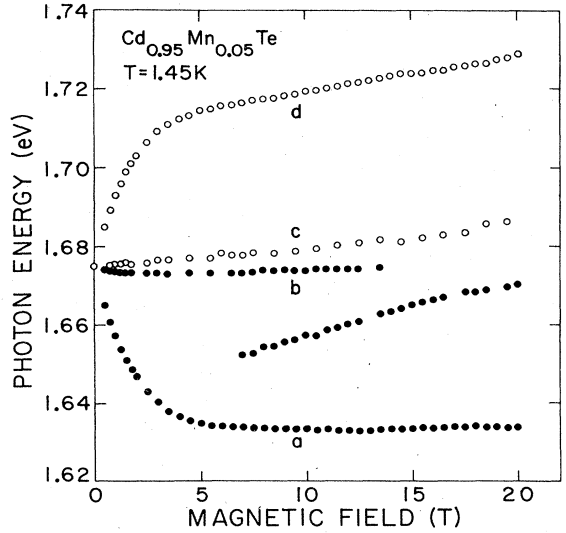

FIG. 2. Magnetic field dependence of the energies of the Zeeman split components a, b, c, and d of the $1s$ exciton in $Cd_{0.95}Mn_{0.05}Te$ at 1.45 K, observed in magnetoreflectance in the Faraday configuration with $\mathbf{B}\|[110]$. Solid (●) and open (○) circles denote the transitions observed for σ_+ and σ_- polarization, respectively. Also shown is an unidentified transition observed, for σ_+ polarization and $B>5$ T.

IV. RESULTS AND DISCUSSION

Figure 2 shows a plot of the observed energies for transitions a, b, c, and d as a function of B for $Cd_{0.95}Mn_{0.05}Te$ at 1.45 K. As expected, transitions a and b are observed in the σ_+ polarization and transitions c and d are observed in the σ_- polarization. Figure 2 also shows an unidentified transition observed in the σ_+ polarization for B above 5 T.

For present purposes, the most interesting feature in Fig. 2 is the behavior of the a and d transitions: The respective excitons undergo relatively large energy shifts for B up to 5 T, and relatively smaller shifts between 5 and 20 T. The midpoint between the a and d branches is also seen to shift to higher energy with increasing field,

due to the diamagnetic energy contribution.

The obscuring effect of the diamagnetic energy shift can be eliminated by plotting the energy difference $\Delta E_{3/2}=E_d-E_a$, as shown in Fig. 3. Using the selection rules and Eqs. (2) and (12), we obtain

$$\Delta E_{3/2}=-x\langle S_z\rangle N_0(\alpha-\beta)+(g_e+6\tilde{\kappa})\mu_B B \ . \quad (16)$$

The contribution of the second term in Eq. (16) due to the conventional Zeeman splitting is estimated from our CdTe data to be ~ -0.08 meV/T. Even at $B=20$ T, this term would contribute only -1.6 meV, compared to the total observed splitting of 95 meV in $Cd_{0.95}Mn_{0.05}Te$. Therefore, we have neglected this contribution in the following analysis by setting

$$\Delta E_{3/2}\simeq -x\langle S_z\rangle N_0(\alpha-\beta) \ . \quad (17)$$

We note further that given $g_e(\mathrm{CdTe})=1.59$,[13] our CdTe data implies $\tilde{\kappa}\simeq 0.04$, which is considerably smaller than the theoretical value of $\tilde{\kappa}=0.93$ obtained from Eq. (15) of Ref. 8 (ignoring the **k** linear term) with valence-band parameters given by Lawaetz.[14]

The solid line in Fig. 3 was obtained as the least-squares fit of the data to Eqs. (10) and (17). The dashed curve is a continuation of the best fit between 0 and 7 T, using only the first term (the Brillouin function) in Eq. (10) for $\langle S_z\rangle$ with the value $\bar{x}/x=0.604$, as obtained previously[2,4] for $Cd_{0.95}Mn_{0.05}Se$, and the fitting parameter $T_0=2.04$ K. Against this reference level, a temperature-broadened step in the exciton splitting is clearly visible between 8 and 15 T. In fact, part of the second step can also be seen at the high-field extreme.

The exchange integrals $N_0\alpha$ and $N_0\beta$ may also be deduced from data in Fig. 2; using $\bar{x}/x=0.604$ and the Brillouin function saturation value of 85.0 meV for $\Delta E_{3/2}$, we obtain $N_0(\alpha-\beta)=1.126$ eV. We further obtain the ratio $\beta/\alpha=-3.7\pm0.2$ by comparing $\Delta E_{3/2}$ with $\Delta E_{1/2}$ in the range 0—7 T. Combining these results, we deduce values of $N_0\alpha=0.24\pm0.02$ eV, and

$$N_0\beta=-0.89\pm0.05 \text{ eV} \ ,$$

in agreement with the values $N_0\alpha=0.22\pm0.01$ eV and

$$N_0\beta=-0.88\pm0.04 \text{ eV}$$

reported previously.[1]

In order to examine the step structure in Fig. 3 more closely, the difference between $\Delta E_{3/2}$ and the Brillouin portion of the splitting $\Delta E^{\mathcal{B}}_{3/2}$ is plotted in Fig. 4. The solid line in Fig. 4 was generated from the second term in Eq. (10), with $T=1.45$ K (the measured sample tempera-

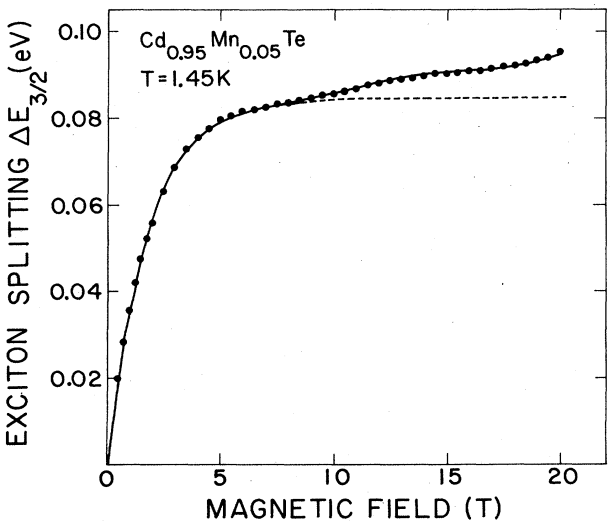

FIG. 3. Magnetic field dependence of the splitting, $\Delta E_{3/2}$, between the d and a components of the $1s$ exciton in $Cd_{0.95}Mn_{0.05}Te$ at 1.45 K. Solid circles (●) show the data points. The solid curve shows the best fit to the data, and the dashed curve shows the contribution of the Brillouin function alone.

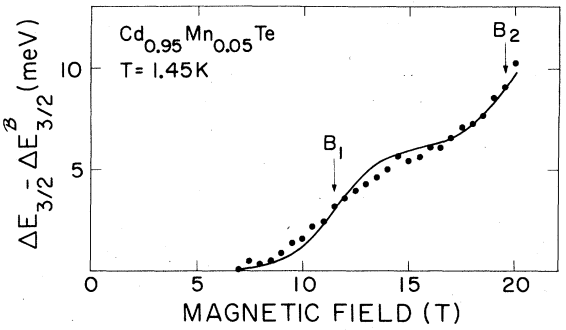

FIG. 4. Magnetic field dependence of the exciton splitting $\Delta E_{3/2}$ (relative to $\Delta E^{\mathcal{B}}_{3/2}$ which denotes the contribution of the Brillouin function) showing steplike behavior attributed to nearest-neighbor (NN) Mn^{2+}-ion pairs in $Cd_{0.95}Mn_{0.05}Te$ at 1.45 K. Solid circles (●) denote the data points. The solid curve is the best fit for the first step $B_1=11.5$ T, the second step $B_2=19.5$ T, and the step size of 6.0 meV, corresponding to the probability $P_p=0.21$ for the NN pairs.

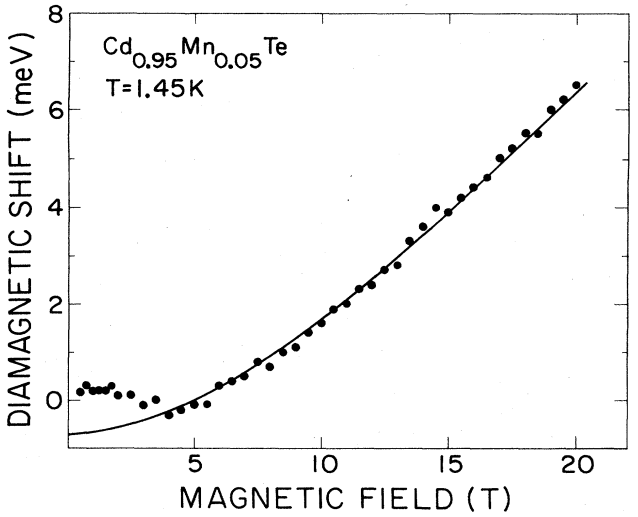

FIG. 5. Diamagnetic energy shift versus magnetic field for the $1s$ exciton in $Cd_{0.05}Mn_{0.05}Te$ at 1.45 K. The solid circles (●) denote the data points. The solid curve was calculated from Larsen's hydrogenic model (Ref. 6), using $\epsilon=10.3$ for the static dielectric constant, and $\mu=0.073m_0$ for the reduced electron-hole mass. The solid curve as shown is shifted down by 0.7 meV.

ture) and the parameters P_p, B_1, and B_2 adjusted for the best overall fit. Parameter values for this fit were found to be $P_p=0.21$ (corresponding to step height of 6.0 meV in Fig. 4), $B_1=11.5$ T, and $B_2=19.5$ T. The above value of 0.21 for P_p is close to the value of 0.24 calculated for a random distribution of Mn^{2+} ions. Substituting $B_1=11.5\pm0.5$ T, and $g_{Mn}=2.0$ in Eq. (6), we obtain $J_{NN}=-7.7\pm0.3$ K. It is interesting to note that the observed value of $B_2=19.5\pm1.0$ T is below the expected value $B_2=2B_1=23.0\pm1.0$ T. The above discrepancy may be due to neglect of the pair interaction with the second-nearest neighbors. Preliminary calculations[15] indicate that J_{NN} should be obtained from the following equation:

$$J_{NN}=-g_{Mn}\mu_B(B_2-B_1)/2\ , \tag{18}$$

if the interaction with the second-nearest neighbors is important. This would lead to a somewhat lower value for $|J_{NN}|$. However, a confirmation of this conjecture will have to await further experimental and theoretical work.

It should be noted that our results differ in two important respects from those of Galazka, Nagata, and Keesom[16] who, on the basis of low-temperature specific heat and magnetic-susceptibility measurements, deduced $J_{NN}=0.55\pm0.05$ K and a strong departure from random distribution of Mn^{2+} ions in $Cd_{1-x}Mn_xTe$ for $0.002<x<0.17$. We believe that at these low concentrations of Mn^{2+} ions, Galazka *et al.*[16] were largely observing the effects due to higher-order neighbors; this would account both for the much smaller value of $|J_{NN}|$ and the apparently larger concentration of pairs than that predicted for a random distribution. Our value for J_{NN} is in much better agreement with the value $J_{NN}=-6$ K obtained from inelastic neutron scattering[17] in $Cd_{0.35}Mn_{0.65}Te$, the latter being consistent with specific-heat measurements in $Cd_{0.5}Mn_{0.5}Te$.[18]

The diamagnetic energy of the exciton is obtained by subtracting zero-field energy from the mean energy of the a and d transitions. Magnetic field dependence of the diamagnetic energy of the exciton obtained in this manner is shown in Fig. 5. The solid curve superimposed upon the data shows the calculated behavior obtained from Larsen's[6] variational results, using $\epsilon=10.3$,[19] and $\mu=0.073m_0$, with resulting $\mathscr{R}=9.4$ meV. To obtain the best fit in the high-field range, however, it was necessary to shift the theoretical curve down by 0.7 meV. We note that the discrepancy between experiment and theory is at its worst in the range 0—5 T, which is precisely the range where exchange effects vary most rapidly with field.

For comparison, Fig. 6 shows the magnetic field dependence of the exciton in CdTe. The best fit of the data with theory is obtained for the same value of $\mu=0.073m_0$ as in the case of $Cd_{0.95}Mn_{0.05}Te$. However, no shift of the theoretical curve was needed. Using the value[20] $m_e=0.0963m_0$ for the effective mass of the conduction electron measured in CdTe and our value of $\mu=0.073m_0$, we obtain $m_h=0.3m_0$ for the effective mass of the hole. This value of m_h may be identified with that of the $M_h=\pm\frac{3}{2}$ (heavy hole) valence band.

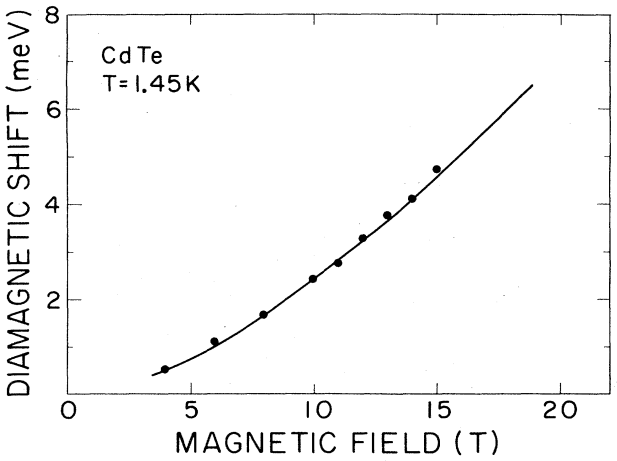

FIG. 6. Diamagnetic energy shift versus magnetic field for the $1s$ exciton in CdTe at 1.45 K. The solid circles (●) denote the data points. The solid curve was calculated from Larsen's hydrogenic model (Ref. 6), using $\epsilon=10.3$ for the static dielectric constant, and $\mu=0.073m_0$ for the reduced electron-hole mass.

In conclusion, we have shown that excitonic magnetoreflectance studies in high magnetic fields provide a direct method for determining the antiferromagnetic exchange interaction between Mn^{2+} ions, in addition to determining the exchange interaction between Mn^{2+} ions and the band-edge electrons and holes.

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