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

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Zeeman splittings of the free exciton in $Zn_{0.95}Mn_{0.05}Te$ have been measured at 1.4 K with dc magnetic field B up to 19.3 T, using magnetoreflectance in the Faraday configuration. The splitting between the highest- and lowest-energy Zeeman components of the exciton follows a modified Brillouin function at low values of $B \lesssim 10$ T. The first of the expected five steps due to the magnetic-field-induced alignment of the antiferromagnetically coupled nearest-neighbor (NN) pairs was observed at $B_1 = 15.0 \pm 0.6$ T. Ignoring the effect of internal fields due to distant-neighbor Mn^{2+} ions on NN pairs, we deduce the NN exchange constant $J_{NN} \simeq -\frac{1}{2} g \mu_B B_1 = -10.1 \pm 0.4$ K, in good agreement with that obtained from magnetization measurements.

I. INTRODUCTION

In this Brief Report we present the results for the optical determination of the nearest-neighbor (NN) Mn^{2+}-Mn^{2+} antiferromagnetic (AF) exchange constant J_{NN} in $Zn_{0.95}Mn_{0.05}Te$, using the Zeeman splitting of the free exciton. This work was motivated by the discrepancy in the values of J_{NN} obtained from the magnetization measurements of Shapira *et al.*[1] and the inelastic neutron scattering measurements of Corliss *et al.*[2] Our optical measurements yield a value of J_{NN} in excellent agreement with that obtained from the magnetization measurements.

The present work on $Zn_{0.95}Mn_{0.05}Te$ follows closely our previous work on $Cd_{0.95}Mn_{0.05}Te$,[3] which also has the zinc-blende structure, and makes use of the theoretical background developed in that paper.

II. EXPERIMENTAL

The single crystal of $Zn_{0.95}Mn_{0.05}Te$ was grown using a modified Bridgman technique. The sample was cut, polished, then etched for approximately 1 min in a 5% solution of bromine in methanol. Magnetoreflectance measurements were made in the Faraday configuration with the sample immersed in liquid helium at 1.4 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 up to 19.3 T. The optical spectrometer employed a 0.22-m double-grating monochromator (Spex model 1680B) equipped with 0.5-μm blazed reflection gratings having 1200 grooves/mm, a tungsten halogen lamp (Sylvania type No. 2097), a long-wave pass filter (Corning Glass No. CS3-73), a circular sheet polarizer (Polaroid model No. HNCP) with design wavelength of 0.56μm, a 150-Hz light beam chopper, a silicon photodiode detector (EG&G model No. SGD-444), and a lock-in amplifier (PARC model No. 5101). The magnetoreflectance spectra were recorded using 0.3-mm-wide monochromator slits.

III. RESULTS AND DISCUSSION

Figure 1 shows a plot of the transition energies for the four Zeeman components observed in magnetoreflectance spectra of $Zn_{0.95}Mn_{0.05}Te$ at 1.4 K. The components a and b are observed in the σ_+ polarization, and the components c and d are observed in the σ_- polarization. The labeling convention is the same as in Ref. 3.

The large splitting $\Delta E_{3/2} \equiv E_d - E_a$ is due to the exchange interaction between the Mn^{2+} ions and the band electrons, and can be written as[3]

$$\Delta E_{3/2} \simeq -x \langle S_z \rangle N_0(\alpha - \beta) , \quad (1)$$

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+} spin, and $N_0\alpha$ and $N_0\beta$ are the exchange constants between Mn^{2+} ions and conduction- and valence-band electrons, respectively. In writing Eq. (1), we have neglected the relatively small conventional Zeeman splitting.

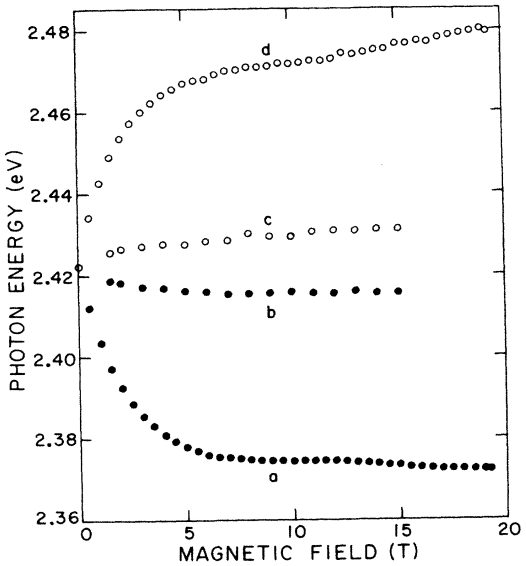

FIG. 1. Magnetic field dependence of the energies of the Zeeman split components a, b, c, and d of the $1s$ exciton in $Zn_{0.95}Mn_{0.05}Te$ at 1.4 K, observed in magnetoreflectance in the Faraday configuration. Solid (●) and open (○) circles denote the transitions observed for σ_+ and σ_- polarization, respectively. The data for a and d transitions were obtained from measurements in a 20-T magnet; data for b and c transitions were obtained from separate measurements in a 15-T magnet.

The average z component of the Mn^{2+} spin can be written as

$$\langle S_z \rangle = -\frac{5}{2}\left[\frac{\bar{x}}{x}\right]\mathcal{B}_{5/2}\left[\frac{5g_{Mn}\mu_B B}{2k(T+T_0)}\right] - \frac{1}{2}P_p\left[1+\exp\left[\frac{g_{Mn}\mu_B}{kT}(B_1-B)\right]\right]^{-1}, \quad (2)$$

where $\bar{x}$ represents the effective molar concentration of isolated Mn^{2+} ions, $\mathcal{B}_{5/2}$ is the Brillouin function of index $\frac{5}{2}$, T_0 is the phenomenological parameter characterizing the AF interaction of the relatively isolated Mn^{2+} ions with distant neighbors, P_p is the probability for the presence of NN Mn^{2+} ion pairs, and B_1 is the field value for the first magnetization step. The first term in Eq. (2) represents the contribution of the isolated Mn^{2+} ions and Mn^{2+}-ion clusters excluding pairs. The second term in Eq. (2) represents the first step arising from the NN pairs; the contributions of the remaining four steps are not included because they lie outside the field range of this experiment.

The magnetic field dependence of $\Delta E_{3/2}$ is shown in Fig. 2. The solid circles represent data points and the solid curve represents the least-squares fit to Eq. (1). The dashed curve is a continuation of the best fit between 0 and 12 T, using only the first term (the Brillouin function) in Eq. (2) for $\langle S_z \rangle$, with $g_{Mn}=2.0$, $\bar{x}/x=0.604$ as obtained previously for $Cd_{0.95}Mn_{0.05}Se$,[4,5] and the fitting parameter $T_0=2.61$ K. Relative to this reference level, a temperature-broadened step in $\Delta E_{3/2}$ is clearly evident between 12 and 19 T.

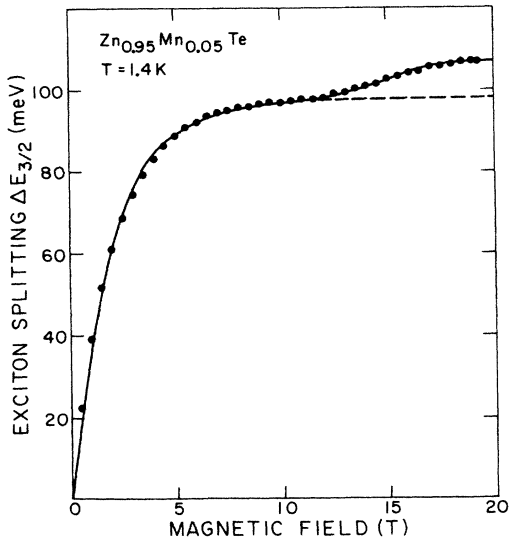

FIG. 2. Magnetic field dependence of the splitting, $\Delta E_{3/2}$, between the d and a components of the $1s$ exciton in $Zn_{0.95}Mn_{0.05}Te$ at 1.4 K. Solid circles (●) show the data points. The solid curve shows the best fit to the data, and the dashed curve shows the contribution of the Brillouin function alone.

The values for the exchange integrals $N_0\alpha$ and $N_0\beta$ may also be obtained from Figs. 1 and 2. Using the best-fit Brillouin function saturation value of 98.2 meV for $\Delta E_{3/2}$, we obtain $N_0(\alpha-\beta)=1.30$ eV. By comparing $\Delta E_{3/2}$ with $\Delta E_{1/2}\equiv E_c-E_b=14\pm1$ meV, we deduce the ratio $\beta/\alpha=-6.0\pm0.4$. Combining these results, we obtain $N_0\alpha=0.19\pm0.02$ eV, and $N_0\beta=-1.11\pm0.05$ eV, in excellent agreement with previously reported values.[6]

In order to highlight the step contribution of the Mn^{2+} pairs to $\Delta E_{3/2}$, the difference between $\Delta E_{3/2}$ and the calculated Brillouin portion of the splitting $\Delta E^{\mathcal{B}}_{3/2}$ is plotted in Fig. 3. The solid line in Fig. 3 represents the best fit,

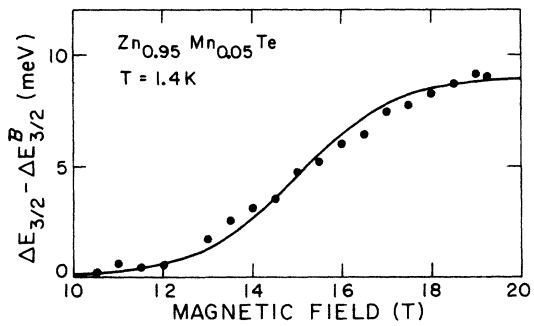

FIG. 3. Magnetic field dependence of the exciton splitting $\Delta E_{3/2}$ (relative to $\Delta E^{\mathcal{B}}_{3/2}$, which denotes the contribution of the Brillouin function) showing steplike behavior attributed to nearest-neighbor (NN) Mn^{2+} ions pairs in $Zn_{0.95}Mn_{0.05}Te$ at 1.4 K. Solid circles (●) denote the data points. The solid curve is the best fit for the first step $B_1=15.0$ T and step size of 9.0 meV, corresponding to the probability $P_p=0.27$ for the NN pairs.

using the second term in Eq. (2) with $T=1.40$ K (the measured sample temperature), and corresponds to $P_p=0.27$ and $B_1=15.0$ T.

The above value of P_p (corresponding to the observed step height of 9.0 meV) is somewhat higher than the value of 0.24 calculated for a random distribution of Mn^{2+} ions in a sample with $x=0.05$; $P_p=0.24$ would correspond to a step height of 8.0 meV. The apparent discrepancy in the step height may be due to the conventional Zeeman splitting neglected in this analysis and/or to the uncertainty in x ($x=0.05\pm0.002$). However, the step position B_1 is affected only slightly by this uncertainty in the step height; B_1 changes by approximately 0.1 T per 1 meV change in the step height. We estimate the uncertainty in B_1 from all sources to be ±0.6 T.

In the absence of internal fields due to distant neighbors, the exchange constant J_{NN} for Mn^{2+} NN pairs is given by[3,4]

$$J_{NN}=-\tfrac{1}{2}g_{Mn}\mu_B B_1 \ . \tag{3}$$

Using the value $B_1=15.0\pm0.6$ T, we obtain $J_{NN}=-10.1\pm0.4$ K. This value is in close agreement with the value $J_{NN}=-10.0\pm0.7$ K determined from magnetization measurements,[1] but well outside the range $J_{NN}=-8.79\pm0.14$ K determined from inelastic neutron scattering measurements.[2]

Recent theoretical work[7] has shown that Eq. (3) overestimates the magnitude of J_{NN}, due to the neglect of internal fields from distant neighbors. For example, the internal field correction reduces $|J_{NN}|$ by 18% in the case of $Cd_{0.95}Mn_{0.05}Te$ and by 9% in the case of $Cd_{0.05}Mn_{0.05}Se$. Thus the 13% apparent discrepancy between the inelastic neutron scattering value of $|J_{NN}|$ and our value of $|J_{NN}|$ could be largely due to the internal field corrections in the two measurements.

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