Oscillatory photoconductivity and photomagnetoresistance in the diluted magnetic semiconductor $Cd_{1-x}Mn_xTe$

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The photoconductivity in the diluted magnetic semiconductor $Cd_{1-x}Mn_xTe$ has been measured between 0.34 and 4.25 K and in magnetic fields up to 8 T. Below the band-gap energy the photocurrent spectra are interpreted in terms of exciton dissociation. Above the band gap the photocurrent oscillates as a function of energy, and the oscillation period suggests an interaction of hot carriers with longitudinal-optical phonons. Due to the large exchange interaction in this material, the photocurrent spectra shift to lower energy in the presence of a magnetic field. The experimental data are in satisfactory agreement with published measurements of magnetoabsorption. The magnetic-field dependence of the photoresistance was studied at a constant wavelength and for various temperatures. Observed oscillations as a function of magnetic field are explained in terms of exchange and optical-phonon interactions.

I. INTRODUCTION

In diluted magnetic semiconductors (DMS) the large exchange interaction between the spins of the free carriers and those of the localized electrons of the magnetic ions gives rise to a large splitting of the conduction and valence bands, leading to many novel physical properties.[1] Due to the difficulty in doping (CdMn)Te, however, the low-temperature transport properties of this material cannot easily be investigated. In particular, it is of interest to study the magnetoresistance in this material and compare it to noncubic DMS, such as (CdMn)Se.[2] Photomagnetoresistance offers a method for probing the transport properties in undoped materials but the interpretation is more complex than normal transport. Only very few papers deal with the effect of a magnetic field on the photoconductivity in (CdMn)Te. We note, for example, the work of Lindström *et al.*,[3] who observed a large negative magnetoresistance which they attributed to a magnetic-field-dependent mobility of the photocarriers.

In this paper we present photoconductivity measurements in $Cd_{0.8}Mn_{0.2}Te$ samples as a function of electric field (up to about 800 $V\,cm^{-1}$), magnetic field (0–8 T), and temperature (0.34–42.5 K). The paper is organized as follows. Section II describes the experimental procedure; Sec. III gives details of the photoconductivity measurements in zero-magnetic field. An excitonic photocurrent is observed and the effects of an electric field are described. Above the band gap, the photocurrent spectra have an oscillatory behavior related to the coupling of hot carriers with LO phonons; Sec. IV discusses the magnetic-field dependence of the photoconductivity. Large red shifts of the photocurrent spectra are observed. Finally, we report oscillatory photomagnetoresistance at a constant wavelength. A study of the temperature dependence of these oscillations demonstrates that the combined effects of the exchange interaction and coupling of hot carriers with longitudinal optical (LO) phonons are responsible for the effect.

II. EXPERIMENT

$Cd_{1-x}Mn_xTe$ single crystals were grown by the vertical Bridgman-Stockbarger method from high-purity materials at the Massachusetts Institute of Technology. The molar-fraction Mn concentration was checked by measuring the high-temperature (100–300 K) Curie constant, $x=0.2\pm0.01$. They were p type with a hole concentration of about 10^{16} cm^{-3} at room temperature. Thin slices of about 1 mm of thickness were cut out of the boule and both sides were mechanically and chemically polished in a 1% solution of bromine methanol. Ohmic contacts were made with HgTe material deposited by vapor-phase epitaxy (2 h at 400 °C), a procedure first suggested by Janick and Triboulet[4] for p-type CdTe. Samples for photocurrent measurements were then cleaved from the slice leading to an active surface exempt of chemical treatment. At the end of the process the illuminated surface was about 0.8×1 mm^2. Copper wires were bound to the Ohmic contacts with silver epoxy. The samples were then placed in a ^3He cryostat with light from a dye laser source guided to the sample surface through an optical cable coupled to an optical fiber. The monochromatic light beam was modulated at a constant frequency of 200 Hz and the photocurrent was measured with a PAR 124 lock-in amplifier.

III. PHOTOCONDUCTIVITY IN ZERO MAGNETIC FIELD

Figure 1 shows the normalized photocurrent near the fundamental absorption edge for various values of the electric field E at a constant temperature of 4.2 K. Also reported in Fig. 1 is the photoluminescence intensity for the same sample, which peaks at $E_{ph}=1.899$ eV. In this latter experiment the excitation source was an Ar laser. For high-Mn concentrations, the photoluminescence is a rather broad line [full width at half maximum (FWHM) of 18 meV] associated with recombination of excitons bound to neutral acceptors. Heiman *et al.*[5] have determined an empirical relation for E_{ph} at 4 K as a function of the molar fraction x; they found, with E_{ph} in meV,

$$E_{ph}=1575+1536x \quad \text{for } 0.2\lesssim x\lesssim 0.4 \ .$$

This relation leads to a value of $x=0.21$, in good agreement with the value extracted from the high-temperature Curie constant. Below the fundamental absorption edge, the photocurrent curves exhibit two lines at, respectively, 1.923 eV and 1.913 eV, labeled E_2 and E_1 in Fig. 1. These lines are interpreted in terms of dissociation of free excitons (E_2) and excitons bound to defects (E_1). The concept of exciton dissociation has also been proposed by Zareba *et al.*[6] to explain their photoconductivity results in $Cd_{0.3}Mn_{0.7}Te$ at 77 K. From the photoluminescence line E_{ph} and the free-exciton peak E_2, we determine a binding energy to the defect, $E_2-E_{ph}=24$ meV. This large value was first measured by Golnick *et al.*[7] and was interpreted in terms of acceptor-bound magnetic-polaron (BMP) formation. Several groups have discussed models for BMP in diluted magnetic semiconductors.[8,9] In photocurrent spectra, which are similar to absorption spectra in many respects, magnetic polaron formation cannot be seen, and as a consequence only the electrostatic part of the binding energy of the exciton-neutral acceptor complex is expected to be measured. For the present experiment the corresponding value of this energy is $E_2-E_1=10$ meV and is close to the value measured in pure CdTe.

We had indicated previously that the E_2 and E_1 lines are due to free and bound excitons. Of course, to make contributions to the photocurrent, excitons have to dissociate. In addition to mechanisms such as interactions with phonons, impurity centers, or excitons, the exciton dissociation may have its origin in the applied electric field E. As a proof for this, we have plotted in Fig. 2 the relative photocurrent $I_r=I_{ph}/I_{ph}(E\to 0)$, where I_{ph} is the photocurrent intensity for the E_2 and E_1 lines as a function of E. The relative photoluminescence intensity $I_{rL}=I_L/I_L(E\to 0)$, where I_L is the area under the photoluminescence peak, is also shown in Fig. 2. The increase in I_r and corresponding decrease in I_{rL} for electric fields higher than 50 V cm^{-1} clearly demonstrates that the electric field participates in the exciton dissociation. Impact ionization by field-accelerated electrons[10] is the most probable mechanism.

Beyond the excitonic region, i.e., for photon energies higher than about 1.930 eV, the photocurrent curves exhibit an oscillatory behavior which is most pronounced at

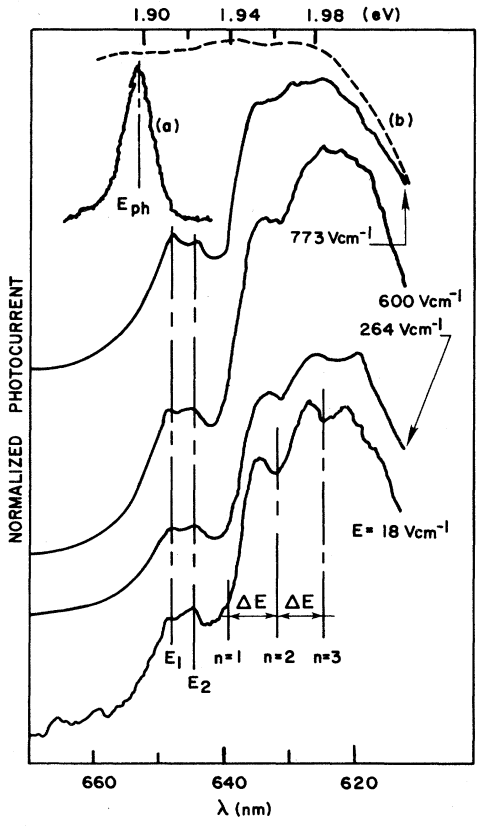

FIG. 1. Normalized photocurrent spectra in zero-magnetic field at 4.2 K for various values of the electric field E in $Cd_{0.8}Mn_{0.2}Te$. The photoluminescence spectrum is in the upper left corner, curve (a). The dye-laser power curve is the dashed line, curve (b).

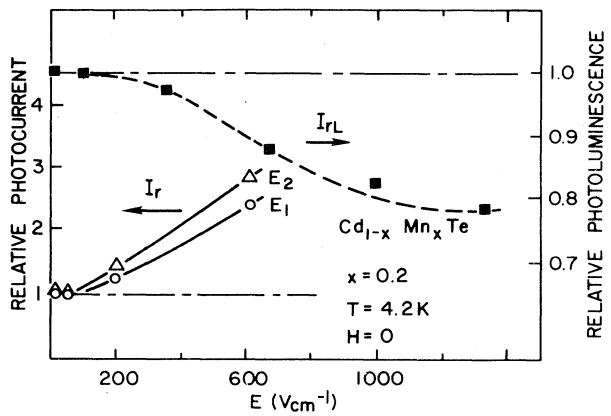

FIG. 2. Relative photocurrent I_r of excitonic lines E_1 and E_2 (solid lines) and relative photoluminescence intensity I_{rL} (dashed line) as a function of the electric field E at 4.2 K in $Cd_{0.8}Mn_{0.2}Te$.

low values of the electric field. To the best of our knowledge, this effect has not been reported in $Cd_{1-x}Mn_xTe$ although many papers deal with similar observations in pure CdTe.[11–14] Oscillatory photoconductivity has also been observed in other semiconductors and is a consequence of the strong interaction of photoexcited carriers or excitons with LO phonons. The oscillation period, ΔE, measured between photocurrent minima, labeled $n=1,2,3,\ldots$, in Fig. 1 is about 23 meV and is larger than the longitudinal-optical-phonon energy determined by Raman scattering.[15,16] Although Raman scattering in mixed crystal $Cd_{1-x}Mn_xTe$ shows "two-mode" behavior corresponding to the "CdTe-like" ($\hbar\omega_0=20$ meV) and "MnTe-like" ($\hbar\omega_0=24.5$ meV) LO modes, we may assume that in an 80 at.% Cd-concentration sample most of the interactions of hot carriers occur through the CdTe-like LO modes. The creation of hot excitons relaxing by emission of LO phonons would lead to a period equal to $\hbar\omega_0=20$ meV which is inconsistent with our results. However, the relaxation of hot electrons (or holes) to the bottom of the conduction band (or the top of the valence band) are known to produce photocurrent minima at energies[14]

$$(h\nu)_m = E_g + n\hbar\omega_0(1+m_e/m_h)\ . \tag{1}$$

The energy E_g is the collector level equal to the forbidden-gap energy, n is an integer, and m_e/m_h is the ratio-electron and hole-effective masses. The factor $(1+m_e/m_h)$ comes from the curvature of the bands.

The period ΔE yields $m_e/m_h=0.15$, a value very close to that for pure CdTe, i.e., $m_e/m_h=0.16$ with $m_e=0.1m_0$ and $m_h=0.6m_0$.[17] Moreover, according to Eq. 1, period ΔE suggests that the photoconductivity is principally due to electrons. Oscillatory photoconductivity in CdTe is not well understood and various results have been reported. For example, in Ref. 11 the photocurrent maxima in the band-to-band region are attributed to the dissociation of excitons generated by a photon accompanied by a simultaneous emission of a phonon. The period of the maxima is said to equal the energy of one longitudinal-optical phonon. It is also observed that oscillatory photoconductivity is surface-state dependent. In Ref. 12 the measured period in n-type CdTe is close to the LO phonon energy $\hbar\omega_0$, whereas in Refs. 13 and 14 the period is higher than $\hbar\omega_0$, with the oscillations also being surface-state dependent.[13]

Our measurements on $Cd_{0.8}Mn_{0.2}Te$ show that the collector level estimated at 1.917 eV lies below E_g, in the forbidden-band-gap energy between the E_1 and E_2 lines. Similar observations were made by Vanecek *et al.*[14] in pure CdTe. They assumed that hot electrons (or holes) relax not only to the bottom (or top) of the conduction (valence) band but also to ground and higher exciton states. In our $Cd_{0.8}Mn_{0.2}Te$ samples, it is presumed that the relaxation is to excited states of the bound exciton. Finally, at higher electric fields the oscillations tend to vanish because the monoenergetic distribution of carriers in the bands is spread out.[18]

IV. PHOTOCONDUCTIVITY IN THE PRESENCE OF A MAGNETIC FIELD

Figure 3 shows the normalized photocurrent spectra as a function of the wavelength at a constant temperature of 4.2 K and a constant value of the electric field $E=200$ V cm^{-1} for various values of the magnetic field B up to 8 T. Linearly polarized light in the Faraday configuration was used in this experiment. As the magnetic-field intensity is increased, the photocurrent spectra shift towards lower energy with a relative decrease of the E_1 line intensity compared to the E_2 line, and the exciton lines are broadened. The large exchange interaction in this material is responsible for the shift, and according to magneto-optical and magnetization studies performed on $Cd_{1-x}Mn_xTe$, the exciton shift corresponding to the lowest transition in σ^+ polarization is[19]

$$|\tfrac{3}{2},-\tfrac{3}{2}\rangle \rightarrow |\tfrac{1}{2},-\tfrac{1}{2}\rangle = (\alpha N_0+\beta N_0)x\langle S_z\rangle/2\ . \tag{2}$$

Here αN_0 and βN_0 are the exchange integrals for the conduction and the valence bands which have been evaluated by Gaj *et al.*[20] to be $\alpha N_0=220\pm10$ meV and $\beta N_0=880\pm40$ meV, x is the Mn molar fraction, and

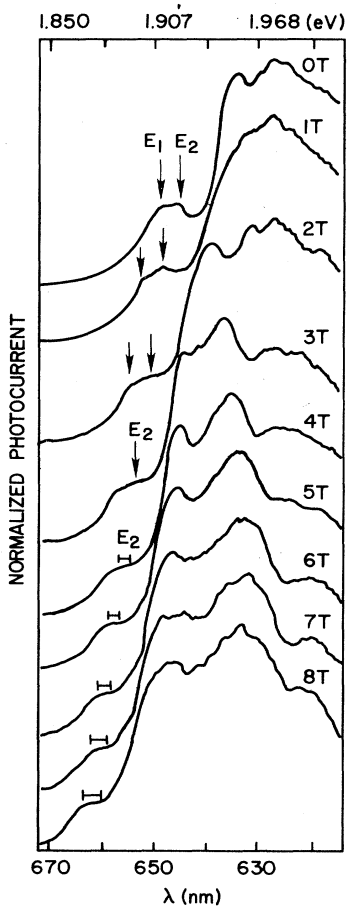

FIG. 3. Normalized photocurrent spectra at constant electric field $E=200$ V cm^{-1} in $Cd_{0.8}Mn_{0.2}Te$ at 4.2 K and for various values of the magnetic field.

$\langle S_z \rangle$ is the Mn^{2+} mean spin value in the field direction. The latter can be evaluated directly from magnetization measurements through the following approximation:[20]

$$\langle S_z \rangle = S_M \beta_{5/2}[\tfrac{5}{2} g\mu_B / k(T+T_0)] , \quad (3)$$

with S_M the Mn^{2+} spin saturation value and $\beta_{5/2}$ the modified Brillouin function. g is the Landé factor, μ_B the Bohr magneton, T the temperature and T_0 an effective temperature which reflects the antiferromagnetic interaction among the Mn^{2+} ions. S_M is smaller than $\frac{5}{2}$ because the antiferromagnetic interaction prevents complete spin alignment. For $x=0.2$, S_M and T_0 take the respective values 0.71 and 7.3.

As shown in Fig. 4 the experimental shift of the E_2 line is smaller than predicted by Eqs. (2) and (3), even taking into account the large errors above 4 T, and the discrepancy increases with the increasing magnetic field. A possible explanation for this discrepancy may be the competition between Landau and excitonic levels. At high values of the magnetic field the cyclotron energy $\hbar\omega_c$ (9.5 meV for $B=8$ T) becomes comparable to the dissociation energy E_d of an exciton [$E_d = 13.57 m_{eq}/\epsilon^2$ in eV, with the dielectric constant $\epsilon=10$ and $m_{eq} = m_e m_h/(m_e+m_h)$] and consequently the dissociation of the 1s exciton ground state becomes less probable. We propose, therefore, that the 2s excited state of the exciton contributes to the photocurrent. In this connection we note that absorption lines of the 2s and 3s exciton states of $Cd_{1-x}Mn_xTe$ ($x \lesssim 0.1$) have been clearly observed by Twardowsky *et al.*[21] in excitonic magnetoabsorption. In Fig. 4 we also plot (dashed line) the magnetic field shift for the 2s exciton state using the computations by Praddaude[22] for a hydrogenlike system in a B field. The experimental data clearly lie between the 1s and 2s exciton states, which suggests that the broadened E_2 line is due to the dissociation of both 1s and 2s states.

As can be seen Fig. 3, the E_1 line, which is due to an exciton bound to a neutral acceptor (Sec. III) vanishes with increasing B. We have no explanation for this behavior. Recent results of the magnetic-field dependence of exciton photoluminescence in a 10 at. % Mn concentration sample[5] clearly indicate two lines, one due to the free-exciton recombination and a second due to excitons bound to acceptor impurities. The presence of a magnetic field weakens the second line and the authors suggest that the magnetic field lifts the degeneracy of the two-hole state of the exciton-neutral acceptor complex, leading to a reduction of the binding energy and dissociation of the exciton from the impurity. A similar mechanism may be affecting the E_1 line.

Finally, we present in Figs. 5(a) and 6 a set of magnetoresistance curves [$\Delta\rho/\rho = 1 - I_{ph}/I_{ph}(B \to 0)$] at a constant wavelength of 632.8 nm (He-Ne laser source) and

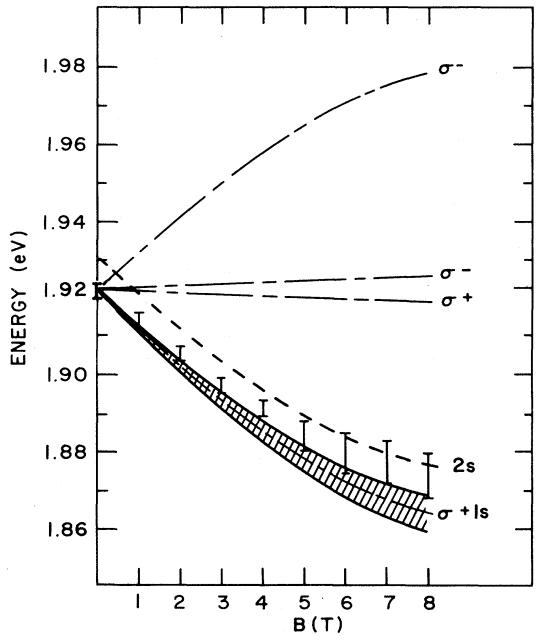

FIG. 4. Experimental shift (vertical lines) and calculated 1s- and 2s-state energy shifts vs magnetic field intensity at 4.2 K in $Cd_{0.8}Mn_{0.2}Te$.

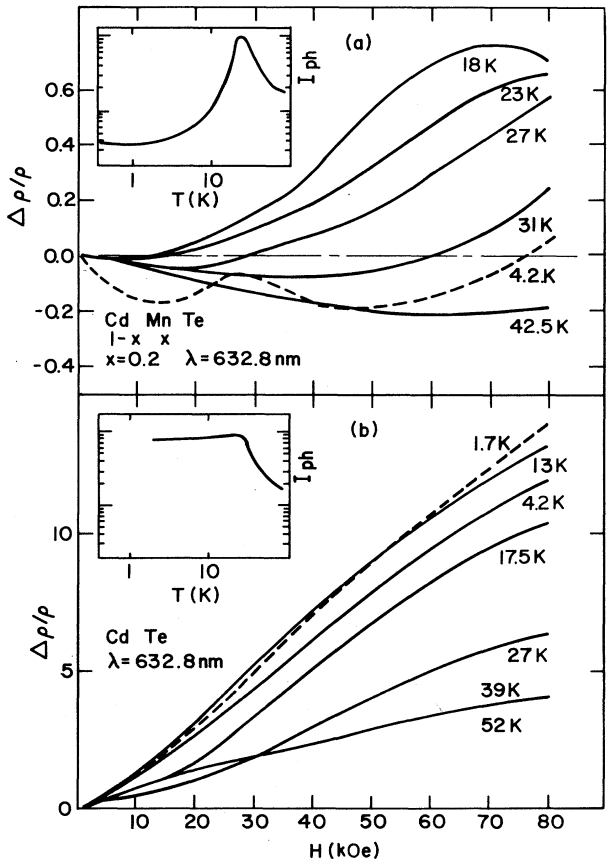

FIG. 5. Photomagnetoresistance $\Delta\rho/\rho$ of (a) $Cd_{0.8}Mn_{0.2}Te$ and (b) pure CdTe as a function of magnetic field at a constant wavelength of 632.8 nm and for various temperatures. Insets in (a) and (b) show the photocurrent vs temperature in zero-magnetic field.

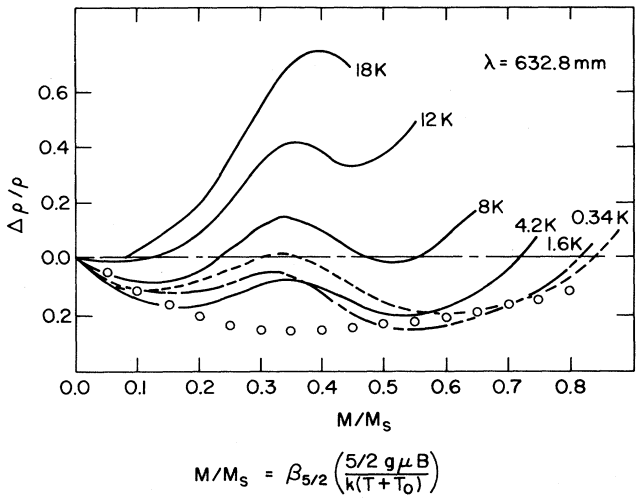

FIG. 6. Photomagnetoresistance $\Delta\rho/\rho$ vs relative magnetization M/M_s at a constant wavelength of 632.8 nm and for various temperatures in $Cd_{0.8}Mn_{0.2}Te$. The open circles represent the photomagnetoresistance measured with a quartz lamp.

for various temperatures from 0.34 to 42 K in a bias electric field $E=200$ V cm^{-1}. Positive and negative contributions are observed. The positive magnetoresistance due to the Lorentz force remains very weak in $Cd_{0.8}Mn_{0.2}Te$ compared to pure CdTe [see Fig. 5(b)]. As seen in Sec. III, the photocurrent in $Cd_{0.8}Mn_{0.2}Te$ is principally supported by one kind of carrier, the electrons. A small positive magnetoresistance is consequently to be expected, although the mobility of samples with Mn seems lower than that of pure CdTe. In CdTe the $\Delta\rho/\rho$ ratio is large and generally decreases as the temperature increases. By comparison, the positive magnetoresistance in $Cd_{0.8}Mn_{0.2}Te$ has a maximum value at about 20 K, probably corresponding to a maximum mobility at this temperature. This conjecture was investigated further by studying the photocurrent as a function of the temperature at constant wavelength of 632.8 nm and in zero magnetic field. Results are reported in the insets of Figs. 5(a) and 5(b) for $Cd_{0.8}Mn_{0.2}Te$ and CdTe, respectively.

Although photoconductivity does not measure the mobility μ but the quantity $\mu\tau$, where τ is the lifetime of carriers, we have observed that the photocurrent versus temperature in $Cd_{0.8}Mn_{0.2}Te$ exhibits a maximum at about 25 K, in fair agreement with magnetoresistance data in this material. The decrease of the photocurrent at low temperature may be related to an impurity-scattering mechanism, since these materials, although undoped, are p type at room temperature with a concentration close to 10^{16}cm^{-3}. However, a change in the lifetime τ as a function of the temperature cannot be ruled out.

The negative magnetoresistance in $Cd_{0.8}Mn_{0.2}Te$ shown in Figs. 5(a) and 6 can easily be understood by looking at the shape of the photocurrent spectra presented in Figs. 1 and 3. Fundamentally, the oscillatory behavior in Figs. 5(a) and 6 is due to the red-shift of the photocurrent curves in Fig. 3 as a function of applied field and, therefore, the magnetization. For example, at an excitation wavelength $\lambda=632.8$ nm and $B=0$ the photocurrent is at a minimum designated $n=2$ in Fig. 1. With increasing B the photocurrent increases (Fig. 3), reaching a maximum for an energy shift $\sim\Delta E/2$ and another minimum at approximately $\Delta E=23$ meV. This accounts for the first maximum in the photomagnetoresistance, Figs. 5(a) and 6, where small changes in the photocurrent spectrum and Lorentz force have been ignored. For example, at 4.2 K the maximum occurs for $M/M_s=\beta_{5/2}=0.35$, which corresponds to a magnetic-field intensity equal to 2.75 T. For this value of β, Eq. (2) predicts a theoretical shift of 26 meV, and the experimental shift, given by the E_2 line in Fig. 4, is 23 ± 1 meV. As the temperature is increased (decreased) an energy shift equivalent to an oscillation period ΔE will be obtained at a higher (smaller) magnetic-field intensity. Thus, since the energy of the optical phonon and the band gap are independent of the temperature in this range ($0.34<T<18$ K), it is expected that the minima in photocurrent curves will occur at the same magnetization value regardless of the temperature. This hypothesis is confirmed in Fig. 6 where all the $\Delta\rho/\rho$ curves have maxima for approximately the same value of the calculated modified Brillouin function. The x axis in Fig. 6 is the relative magnetization, i.e., $M/M_s=\beta_{5/2}[\frac{5}{2}g\mu B/k(T+T_0)]$, where M and M_s are, respectively, the real and saturation magnetization values and where the modified Brillouin function is defined in Eq. (3). Before concluding, let us return briefly to the negative magnetoresistance. This effect had been reported by Lindström *et al.*[3] in high-Mn-concentration samples using a quartz lamp to excite the carriers. They suggested that a magnetic-field-dependent mobility is responsible for a featureless negative magnetoresistance. We have performed the same experiment at 4.2 K (open circles in Fig. 6) and also observe a negative magnetoresistance without any structure. The absence of the maximum is due to the large bandwidth of the excitation source and the photocurrent increase is principally due to the nonuniformity of the spectral irradiance of the lamp. In our view it is unnecessary to invoke magnetic-field-dependent mobility to explain this negative magnetoresistance.

V. CONCLUSIONS

The photoconductivity of $Cd_{0.8}Mn_{0.2}Te$ single crystals has been studied as a function of electric and magnetic field, and temperature. Below the energy gap, electric-field-dependent exciton dissociation is primarily responsible for the photocurrent. A direct comparison of photoluminescence and photocurrent spectra led to the determination of the binding energy of the exciton-neutral acceptor complex. The value found (24 meV) is in good agreement with the energy of a bound magnetic polaron. Above the gap, the photocurrent spectra exhibit oscillatory behavior. The oscillation period is larger than the CdTe-like longitudinal-optical phonon and suggests a strong interaction of hot carriers with optical phonons. As expected, a high electric field suppresses the oscillatory photoconductivity.

Due to the large exchange interaction between the free carriers and localized d electrons of the Mn^{2+} ions, the photocurrent spectra exhibit a large red shift which is, however, slightly weaker than that measured in magneto-absorption. A possible explanation for this effect is based on the fact that the excitonic photocurrent may be produced by an overlap of ground and first-excited-state wave functions of the excitons.

Finally, the photomagnetoresistance measured at a constant wavelength reveals oscillatory behavior as a function of magnetic field. This is a consequence of (i) the strong coupling of photoexcited hot carriers with LO phonons and (ii) the exchange interaction. We also point out that the negative magnetoresistance observed previously in this material is not due to a magnetic-field-dependent mobility, but has its origin in the energy-dependent photocurrent spectrum and the red shift resulting from the exchange interaction.

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