Optically detected magnetic resonance of $Cd_{0.905}Mn_{0.095}Te$

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Studies of the optically detected magnetic resonance of the semimagnetic semiconductor $Cd_{1-x}Mn_xTe$ ($x=0.095$) are presented. It is shown that for an external magnetic field of about 1.2 T the magnetic resonance of Mn^{2+} is detected optically via changes of the $Cd_{0.905}Mn_{0.095}Te$ "edge emission" which are induced by a decrease of the magnetization of the Mn^{2+} spin system. The sample magnetization influences the splitting of exciton and impurity states. As a result of the Mn^{2+} spin transitions, the emission spectra shift toward higher energies enabling optical detection of the magnetic resonance.

I. INTRODUCTION

Optically detected magnetic resonance (ODMR) has become a useful technique for identification of centers taking part in recombination processes.[1] This stems from the fact that under magnetic-resonance conditions the effectivity of the various recombination paths of a center are changed and, thus, the magnetic-resonance spectrum of a center is connected to its optical emission. In this study the ODMR technique is applied to the semimagnetic semiconductor $Cd_{1-x}Mn_xTe$. Recent reviews on some properties of semimagnetic semiconductors can be found in Refs. 2–4.

For a semimagnetic semiconductor one can expect that the following mechanisms might be responsible for ODMR signals: spin-dependent energy transfer,[5–7] spin-dependent scattering,[8–10] cross relaxation,[5,11] effects due to the presence of a strong nonresonant background,[12,13] and magneto-optical or semimagnetic effects.[14–17] The first four mechanisms will mainly lead to changes of photoluminescence (PL) intensity and their contributions are therefore difficult to separate. Consequently, the dominant mechanism enabling optical detection of the magnetic resonance remains often unknown.[11] In contrast, the magneto-optical mechanism can lead to a different response: in this case the various spin states of the carriers involved in an optical transition are split in a magnetic field and thus their respective luminescence lines shift upon a change of the sample magnetization. Under magnetic-resonance conditions this splitting will become smaller. This will result in a derivativelike line shape of the wavelength dependence of the ODMR signal. Furthermore, the line shape may be asymmetric as also the relative intensities of the various components may change.[15] Such a response is then very characteristic and can allow the identification of the magneto-optical effects as the dominating ODMR mechanism; this is shown to be the case in the current study.

In this paper the results of the optically detected magnetic-resonance experiments performed on $Cd_{1-x}Mn_xTe$ samples with $x=0.095$ are presented. For such a value of x, optimal magneto-optical effects can be observed in an ODMR experiment. For higher Mn concentrations the magnetic phase transition occurs for $T>2$ K, prohibiting the ODMR detection, as verified for a $Cd_{1-x}Mn_xTe$ crystal with $x=0.28$. On the other hand, preliminary ODMR studies as performed on a sample with $x\approx 0.01$ show that a lower Mn concentration results in a much smaller magnitude of the magneto-optical effect.

II. EXPERIMENT AND SAMPLE

The PL was excited with the 514.5-nm line of a cw Ar^+ laser operating with an output power of typically 50–100 mW. To avoid spurious plasma lines, a 514.5-nm interference filter was used. The luminescence was collected from the laser-irradiated side and was dispersed with a high-resolution 1.5-m F/12 monochromator (Jobin-Yvon THR-1500) with a 600-grooves/mm grating blazed at 1.5 μm. It was detected by a liquid-nitrogen-cooled germanium detector (North Coast EO-817) and amplified using the conventional lock-in technique for which the luminescence was on-off modulated at 830 Hz.

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Various emission bands could be selected by inserting optical filters in front of the entrance slit of the monochromator. The ODMR experiments were carried out at a microwave frequency $\nu \approx 35$ GHz (Q band) in the Faraday configuration, using a split-coil superconducting magnet with the direction of the magnetic field parallel to the light. The sample was mounted in a cylindrical TE_{001} cavity with slits for optical access. The helium in the cryostat was pumped to prevent noise due to liquid-helium evaporation, leading to a sample temperature of $T \approx 2$ K. The $Cd_{1-x}Mn_xTe$ sample used in this study was grown by the Bridgman method. It contained nominally 10% of Mn. The exact Mn concentration was determined from an optical reflection study giving $x = 0.095$ with some fluctuation within the sample.

III. RESULTS AND DISCUSSION

The photoluminescence spectrum of the $Cd_{1-x}Mn_xTe$ ($x = 0.095$) crystal is given in Fig. 1. In Fig. 1(a) the full spectrum in the wavelength range 700–1400 nm is given and in Fig. 1(b) a higher-resolution spectrum of the high-energy ("edge emission") part is presented. The spectrum consists of several components. The sharp line at 713.3 nm (1738 meV) is identified as a free-exciton (FE) recombination. The line at 721.8 nm (1718 meV) can be attributed to an acceptor-bound exciton (ABE) with an associated phonon replica at 729.5 nm (1700 meV). This assumption is supported by the fact that in CdTe a line observed at an energy 8 meV below the FE line has been identified by several authors[18–20] as an ABE. It has been observed that this line shifts to 20 meV below the FE absorption peak for $Cd_{0.9}Mn_{0.1}Te$ as a result of the formation of a bound magnetic polaron.[21,22] A PL band at 743.7 nm (1667 meV) is attributed to the superposition of a free-to-bound (free-electron-to-acceptor, FA) transition and shallow-donor-related donor-accepted-pair (DAP) recombination, involving the same acceptor as the FA process. In addition, a DAP band is observed at 840 nm (1476 meV) which shows a weak phonon replica; such replicas are not observed for three deeper, superimposed, DAP bands at 960 nm (1292 meV), 1100 nm (1127 meV), and 1190 nm (1042 meV). These DAP bands lie outside the range 1500–1600 meV in which Misiewicz *et al.*[23] observed DAP bands in $Cd_{0.9}Mn_{0.1}Te$ doped with As, P, Au, and Cu, which are the only characterized bands in this material.

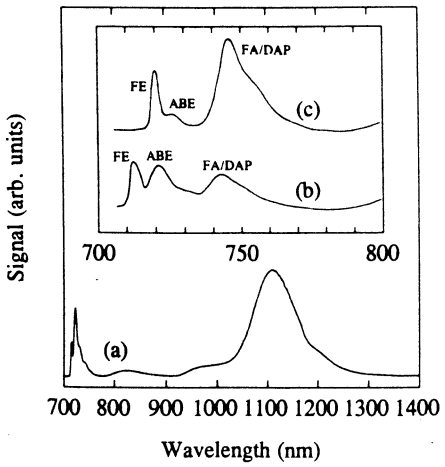

FIG. 1. Photoluminescence spectrum as recorded for the $Cd_{0.905}Mn_{0.095}Te$ crystals. (a) The spectrum between 700- and 1400-nm wavelength as studied at 4.2 K under 100-mW argon laser excitation. (b) The high-energy part of the spectrum in a higher resolution as recorded at 2.0 K under 50-mW laser excitation. (c) Magnetic-field dependence of the "edge" photoluminescence measured at 2.0 K with magnetic field set at 1.2 T.

In the present study none of the observed PL lines could be assigned to an internal Mn^{2+} emission. This is due to the fact that the lowest excited state of Mn^{2+}, its $^4T_1$ state, lies approximately 2.0 eV above its $^6A_1$ ground state. This implies that only for $Cd_{1-x}Mn_xTe$ samples with $x > 0.4$ this excited level is below the conduction-band edge. Consequently, an internal Mn^{2+} emission can be observed only for such crystals, and not in the one studied here.[24,26]

The influence of the magnetic field on the edge emission is shown in Fig. 1(c), where the PL spectrum between 700 and 800 nm at 2 K is given for the magnetic field set at 1.2 T. This field is in the range of the magnetic-resonance peak of a system with $g \approx 2$ in a Q-band spectrometer. As can be seen, when the magnetic field is applied, all emission lines in the edge region shift toward lower energies. As only a shift is observed and no splitting, this indicates that the thermalization is faster than the optical decay rates, and that only one spin state gives rise to the optical emission. This shift can be calculated as 17.9 meV at 2 K for a magnetic field of 1.2 T.[27] This in good agreement with the observed shift of the FE line which is 18.4 meV. In addition to the energy shift, also narrowing of the FE is observed upon the application of the magnetic field.

The magnetic field also has an effect on the line intensities; the intensity of the ABE line is reduced as compared to the intensities of two other lines in the edge region. This may be caused by the valence-band splitting exceeding the binding energy of the exciton on the neutral acceptor; as such an effect has been observed by Planel, Gaj, and Benoit à la Guillaume[28] in a sample with $x = 0.08$ for magnetic fields above 1.25 T. Under such conditions the ABE becomes unstable and may decay into a free exciton and a neutral acceptor in its ground state. This will lead to a decrease in the intensity of the ABE line and an increase in the intensity of the FE and FA/DAP lines, as is observed here. This mechanism cannot, however, fully account for the increase of the FA/DAP band by a factor 3 at 1.2 T as compared to its value at zero magnetic field. All the above features are in good agreement with previously reported properties of FE, ABE, DAP, and FA PL transitions in $Cd_{1-x}Mn_xTe$ with similar x fractions.[3,22,27,29,30]

Figure 2 presents the Q-band magnetic-resonance signal detected as a change of PL intensity for two different wavelengths corresponding to FE and DAP bands. Also, for other wavelengths a similar ODMR signal is record-

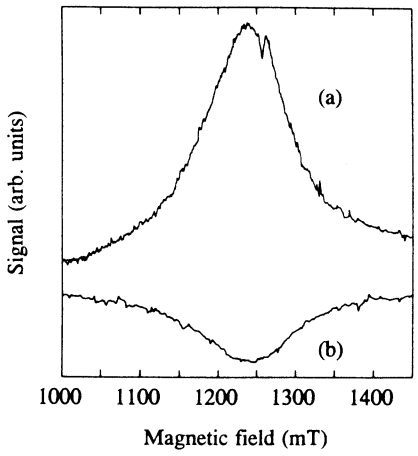

FIG. 2. Optically detected magnetic-resonance spectrum of the $Cd_{0.905}Mn_{0.095}Te$ sample measured as the change of PL intensity for (a) the 719-nm line and (b) the 753-nm line. Measurements were carried out in the Q-band spectrometer ($\nu = 34.8$ GHz) with a microwave power of 230 mW, on-off modulated at 830 Hz.

ed; it is broad and featureless with a g value of 2.00 and a full width at half-maximum of 125 mT. This signal is superimposed on a much weaker nonresonant background. The low intensity of the background signal implies that nonresonant background effects cannot, in our experiment, constitute the dominating ODMR mechanism. We identify the signal as being due to the Mn^{2+} magnetic-resonance transition. A characteristic feature of a manganese resonance, i.e., a sixfold hyperfine splitting, is not observed in this case. This results from the fact that the lines of the Mn spectrum in $Cd_{1-x}Mn_xTe$ broaden with increasing values of x, due to interaction between the various Mn atoms. For values of $x > 0.02$ the lines are completely smeared out and only a broad band is observed.[31–33]

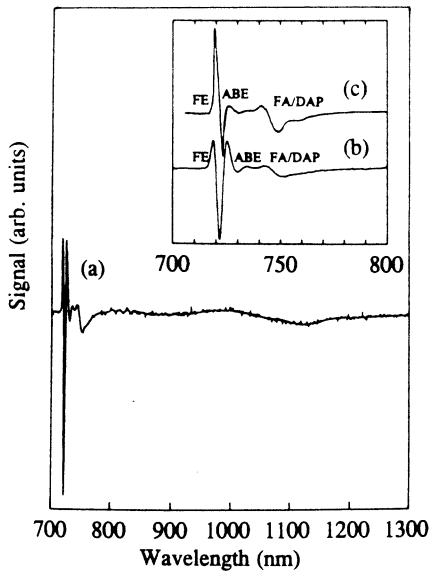

FIG. 3. Spectral dependence of the Mn^{2+} ODMR signal in $Cd_{0.905}Mn_{0.095}Te$. Data were taken at 2.0 K and with the magnetic field set at 1.27 T, i.e., at the maximum of the ODMR signal. Microwave power (230 mW) was on-off modulated at 830 Hz. (a) The full spectrum (700–1300 nm). (b) The high-energy part (700–800 nm). (c) Differential PL spectrum as constructed from the 1.2-T and 1.3-T PL spectra.

The spectral dependence of the ODMR signal (ODMR PL) is shown in Fig. 3, where the response of different PL bands to the Mn^{2+} magnetic resonance is depicted. Figure 3(a) shows the full spectrum between 700 and 1300 nm; in Fig. 3(b) the response of the edge emission is given. The data were taken at 1.27 T, i.e., at the maximum of the ODMR signal. The figure shows that the FA/DAP line has an asymmetric derivativelike spectral dependence and is shifted toward higher energies. The response of the FE and ABE lines overlap, but also for these two lines it can be deduced that they shift toward higher energies under magnetic-resonance conditions. The ODMR PL spectrum of the DAP bands is very weak and is only observed for the strongest one at 1100 nm. Its ODMR PL response is derivativelike and is positioned at ≈ 1065 nm. This band also shows a shift toward higher energies. The shifts of the PL lines indicate that the magneto-optical effect is the dominating ODMR mechanism.

It can also be noticed that the intensity ratio of the various emissions changes upon inducing the magnetic resonance; the intensity of all bands is reduced when compared to the ABE. This agrees with the observation of the magnetic-field dependence of the PL spectrum, where the intensity of the ABE decreased in stronger magnetic fields.

In Fig. 3(c) the difference of the PL spectrum as recorded at 1.2 and 1.3 T [i.e., PL(1.2 T)–PL(1.3 T)] is depicted. This difference resembles the ODMR PL spectrum from Fig. 3(b) thus further supporting the assumption that the reduction of the magnetization in the sample under magnetic-resonance conditions is responsible for the ODMR. It should be noted, however, that the intensity of the response of the ABE line as compared to the other lines is much stronger in the ODMR experiment.

The dependence on microwave power of the ODMR signal was also measured and is shown in Fig. 4(a)–4(c). From this figure it follows that the ratio between the response of the ABE line and the FE line is much smaller for lower microwave powers than for the higher ones. A possible explanation for this is that, as discussed before, the intensity of the ABE line depends very differently on the magnetic-field value (and on the applied microwave power) than the intensity of the FE line. This may lead to a different microwave power dependence of the ODMR PL intensity of the two lines.

Figure 4(d) shows the edge part of the ODMR PL spectrum for the magnetic field set at 1.7 T, i.e., away from the Mn^{2+} resonance peak. The observed spectral dependence is very similar in intensity to the ODMR PL spectrum recorded at 15 mW at the Mn^{2+} resonance field [Fig. 4(c)], with the FE peak shifted toward a lower energy, due to the stronger magnetic field. By comparing the FE shift between 0, 1.3, and 1.7 T one can conclude that

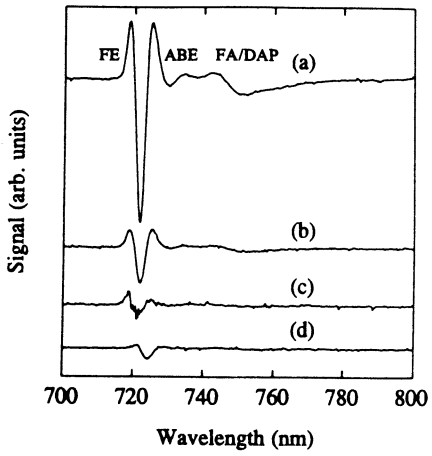

FIG. 4. ODMR PL spectrum of $Cd_{0.905}Mn_{0.095}Te$ as obtained for the magnetic field of 1.27 T at (a) 230-mW, (b) 60-mW, (c) 15-mW microwave excitation, and (d) Spectral dependence of the edge emission measured at 1.7 T synchronously with the *Q*-band microwave power of 230-mW on-off modulated at 830 Hz.

at $T=2$ K no saturation of the magnetization occurs for these values of the magnetic field. The fact that only the response of the FE line is observed is probably caused by the disappearance of the ABE line at 1.7 T. From the observed derivativelike response it follows that also under off-resonance conditions, microwaves lead to a change in the magnetization of the sample. This is most likely due to carrier and lattice heating by the applied microwave power; the spin temperature of the Mn^{2+} spin system increases, thus lowering the magnetization. The change in magnetization under microwave illumination is nearly equal in both cases as the line intensities of the FE peaks in the two ODMR PL spectra are almost identical. The microwave power is 230 mW for the magnetic field at 1.7 T and 15 mW at 1.27 T. This implies that the efficiency of the decrease of magnetization by the microwaves is 15 times higher at 1.27 T (at the maximum of the ODMR signal) than at 1.7 T. One can therefore conclude that the observed effects on the various emission lines at 1.27 T have a strongly resonant character and that lattice and carrier heating have only a small effect.

IV. CONCLUSIONS

Detailed ODMR studies of the $Cd_{1-x}Mn_xTe$ semimagnetic semiconductor are presented. The experimental results allow us to establish the magneto-optical effect as the dominating mechanism for the optical detection of the magnetic resonance at 1.2 T. Under magnetic-resonance conditions the spin temperature of the Mn^{2+} system rises, thus reducing the magnetization. The lower magnetization influences, via direct coupling, the energy levels splitting of the initial as well as the final states involved in the optical transitions. This effect is observed in the ODMR experiment as a derivativelike PL response to the magnetic-resonance transition indicating a shift of the emissions toward higher energies. In addition to the shift, characteristic changes in intensity of the excitonic emissions could be detected.

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