Observation on the high-magnetic-field electron-spin resonance of $Cd_xMn_{1-x}Te$

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Using far-infrared laser pulses, electron-spin resonance has been excited in $Cd_xMn_{1-x}Te$ in high magnetic fields up to 22 T at liquid-helium temperature. Electron-spin-resonance line shapes have been observed by monitoring the transmitted intensity. Simultaneously, the dynamic of the longitudinal magnetization was recorded with a pick-up coil showing, contrary to the transmission, a single line. The discrepancies were due to the geometrical interferences.

I. INTRODUCTION

Recently, many papers have been devoted to electron-spin resonance (ESR) in high magnetic fields.[1–5] The excitation energies corresponding to high-magnetic-field ESR experiments no longer lie in the microwave region as in the case of conventional ESR spectrometers, but in the far-infrared (FIR) part of the electromagnetic radiation spectrum. Therefore, high-field ESR experiments basically involve quasioptical techniques.[6] It has to be pointed out that in high-field ESR experiments—due to the lack of a cavity—magnetic-field and electric-field vectors are simultaneously present at the sample site, which could lead to the quantum interferences between magnetic dipole (MD) and electric dipole (ED) transitions. On the other hand the observed line shapes can be distorted by "geometrical" interference phenomena.

In this paper, we present a comparison of transmission data with dynamic magnetization measurements and discuss the results taking into account geometrical interferences.

Some of the papers mentioned above were devoted to the ESR measurements in diluted magnetic semiconductors[3–5] with manganese. The Mn^{2+} has, in the ground state, a total angular momentum $L=0$ and a total spin quantum number $S=\frac{5}{2}$ which arises from the half-filled $3d$ shell of the ion.[7] States with a total spin quantum number $\frac{3}{2}$ and nonzero angular momentum are excited states. The lowest of these excited Mn states lies about 2 eV higher in energy. The Mn levels are located deeply in the valence band and are weakly hybridized with the latter. $Cd_xMn_{1-x}Te$ belongs to the class of wide-gap semiconductors, having a gap energy of more than 1.6 eV. At liquid-helium temperature no free carriers are present. Therefore, in a first approximation one can consider the Mn^{2+} ground state as a pure magnetic subsystem.

In Ref. 4 the unusual structures in the ESR transmission line shape ("satellite lines") observed for $Cd_{1-x}Mn_xTe$ with $x \leq 0.1$ have been explained as transitions between levels of exchange coupled nearest-neighbor (NN) Mn pairs. It could not explain the increase of transmission in the vicinity of the resonance. To check which part of the structure in transmission is due to transitions between spin states we have measured—simultaneously with the transmission—the changes of the magnetization using a pick-up coil. The results give some new information for the interpretation of the ESR transmission line shapes which are not available in conventional microwave or transmission experiments. This method allows the discrimination between structures in transmission coming from absorption of light due to transitions between spin states (change of magnetization) and coming from other effects (e.g., interferences).

The paper is organized in the following way. First, in Sec. II the experimental details are described and the results discussed. Then, the model for transmission calculations is introduced and possible explanations of experimental results are presented in Sec. III. Finally, in Sec. IV some conclusions are drawn.

II. EXPERIMENTAL DETAILS AND RESULTS

Mixed crystals of $Cd_{1-x}Mn_xTe$ were grown by a modified Bridgman technique at the Institute of Physics, Polish Academy of Sciences. The samples were cut from the ingot with a wire saw and roughly polished in the form of disks with 4-mm diam and thicknesses varying between 0.5 and 1 mm. The Mn concentration was checked using electron microprobe analysis.

Since for measurements of magnetization dynamics pulsed excitations are needed, a powerful and narrow bandwidth FIR optically pumped pulsed molecular FIR laser was used. The FIR laser radiation was sent from the laser to the sample through oversized waveguides.[8] The sample is placed in an external magnetic field, provided either by a 10-T superconducting magnet or a 25-T Bitter-Polyhelix magnet (e.g., Ref. 9). A pick-up coil is placed around the sample with the coil axis parallel to the external static magnetic field. The sample and the pick-up coil are immersed in low-pressure helium gas which is in thermal contact with a liquid-helium bath. Below the sample an Allen-Bradley carbon resistor and a bolometer made out of a similar resistor are fixed for temperature and transmission measurements, respectively. The more detailed description of the setup can be found elsewhere.[8]

A pick-up coil fixed around a paramagnetic sample with its axis parallel to an external magnetic field detects the externally induced changes of the longitudinal magnetization. Under the ESR condition the FIR radiation excites transitions in the Zeeman split energy levels, thus changing the population of the spin energy levels. The changes in population directly lead to changes of the longitudinal magnetization given by the alignment of the Mn^{2+} spins. These changes induce a voltage in the pick-up coil.

For some of the line-shape studies we have also used circular polarized FIR radiation with a wavelength of 496 μm. In this case, a home-made circular polarizer is placed between the end of the waveguide and the sample. It consists of a grid linear polarizer glued on a $\lambda/4$ plate. The $\lambda/4$ plate was made with a properly shaped sapphire crystal. The quality of the polarization obtained in this way was rather poor. In the worst case about 40% of undesired polarization was present.

Typical field sweep spectra of $Cd_xMn_{1-x}Te$ samples with different compositions, as measured with different FIR laser lines, are plotted in Figs. 1 and 2. The two minima seen in transmission [Fig. 1(a), trace a] are due to the ESR absorption at fields corresponding to two laser line energies. Their position leads to a g factor of 2.00 ± 0.01, in agreement with other experiments.[5,10–13] At the same fields a strong signal is induced in the pick-up coil [Fig. 1(a), trace b], which clearly shows that the

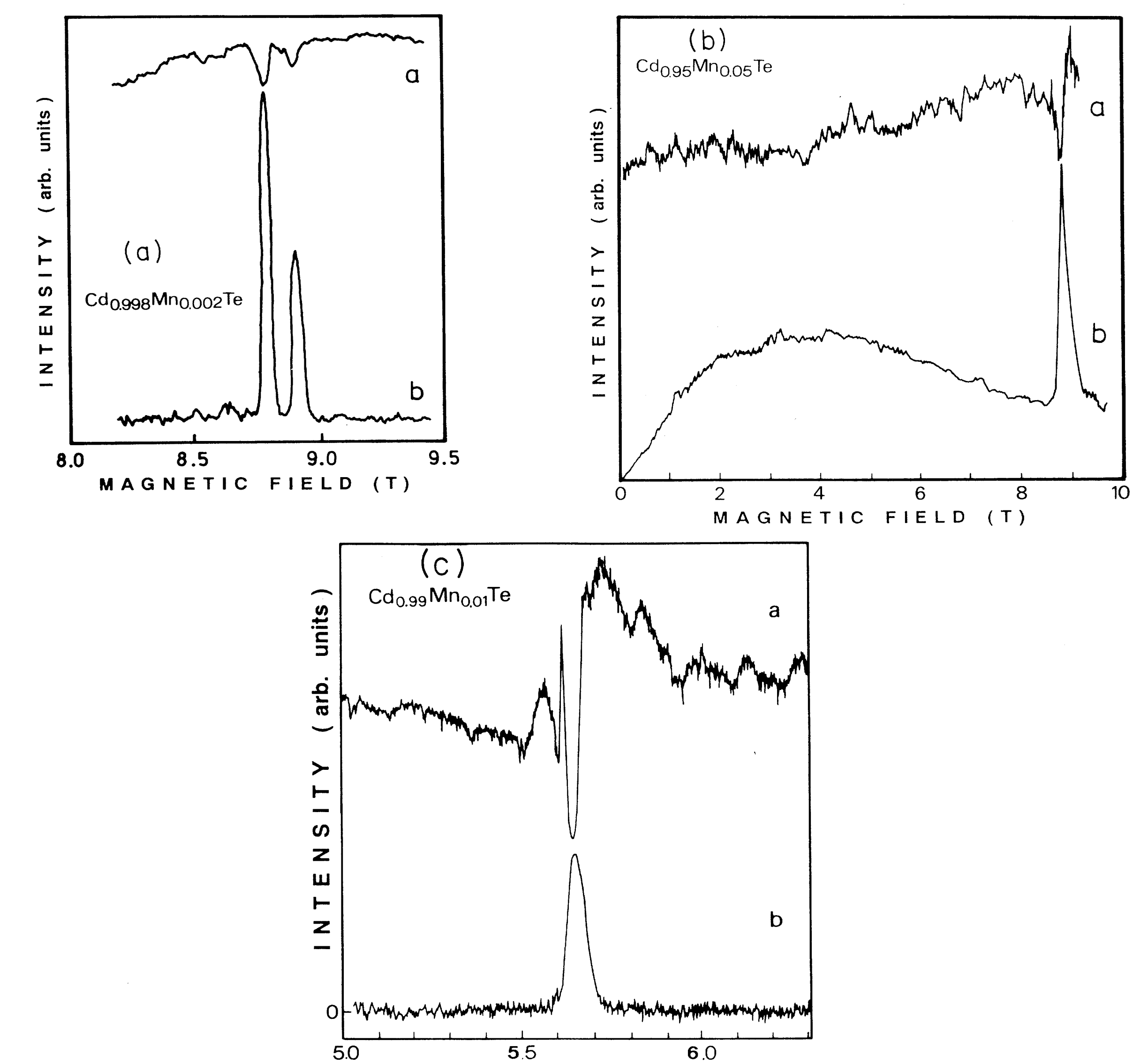

FIG. 1. Transmission (traces a) and induced voltage in the pick-up coil (traces b) in field sweep ESR spectra of $Cd_{0.998}Mn_{0.002}Te$ (a) and $Cd_{0.95}Mn_{0.05}Te$ (b) at 5.2 K for FIR excitation wavelengths of 1207- and 1222-μm lasing, simultaneously. (c) Transmission (line a) and induction signal of the pick-up coil (line b) as a function of the magnetic field for $Cd_{0.99}Mn_{0.01}Te$ at 4.7 K using FIR radiation with a wavelength of 1899.9 μm.

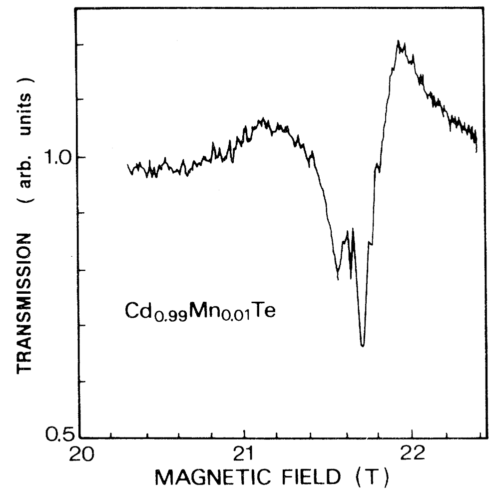

FIG. 2. Transmission of $Cd_{0.99}Mn_{0.01}Te$ at 4.7 K around the ESR field excited with FIR radiation of 496-μm wavelength.

observed pick-up coil signal is due to the magnetization changes caused by ESR absorption. In Fig. 1(b) a spectrum of a $Cd_xMn_{1-x}Te$ sample with a higher Mn concentration, namely $x=0.05$, is shown as measured under the same experimental conditions. Only one ESR line—broader than the line in Fig. 1(a)—can be seen in transmission [Fig. 1(b), trace a]. The transmission line shape is asymmetric and has dispersive character. Again, at the ESR field of about 8.9 T a strong signal (single line) is induced in the coil [Fig. 1(b), trace b]. In the cases presented, the existence of radiation with two different energies does not allow the precise study of transmission line shapes. In Fig. 1(c) the transmission and the coil signal are shown measured for $Cd_{0.99}Mn_{0.01}Te$ at lower fields using a FIR radiation with a wavelength of 1899.9 μm. The transmission curve [Fig. 1(c), trace a] is better resolved and reveals a sharp positive peak in the main absorption line, again of antisymmetric dispersive shape. The coil signal [Fig. 1(c), trace b] does not show these features, but a single maximum at about the magnetic field, where the main transmission minimum occurs. In Fig. 2 the transmission curve for $Cd_{0.99}Mn_{0.01}Te$ is presented as measured around the ESR field of 21.6 T using FIR pulses with wavelengths of 496 μm. It shows more or less the same overall features as the transmission curve of Fig. 1(c), except the broad increase in transmission on the low-field side of the structure.

In Fig. 1(b) one can see a nonzero coil signal at nonzero, but nonresonant magnetic fields. This signal is due to the change of the magnetization caused by heating of the lattice, since the FIR radiation is weakly absorbed by the crystal.[9,14]

III. DISCUSSION

Our results can be compared with other transmission measurements obtained in Faraday configuration at high magnetic fields using unpolarized FIR radiation provided by a continuous-wave FIR laser.[5] In that work similar resonance line shapes were reported. Structures around the main ESR absorption were observed and called "satellite resonances." They were interpreted as absorption due to excitation in exchange coupled NN Mn pairs. However, the increase of transmission around the resonance has not been discussed by the previous authors.[5] Our results cannot fully support this explanation, since we do not observe any change in the pick-up coil signal at fields where the "satellite resonances" occur. Since the pick-up coil is a tool to detect magnetization changes, one would expect to see these "satellite resonances" also in the pick-up coil signal, if they are due to transitions induced between pair energy levels (different spin projections on the quantization axis). Such transitions would change the pair magnetization. Judging from the relative

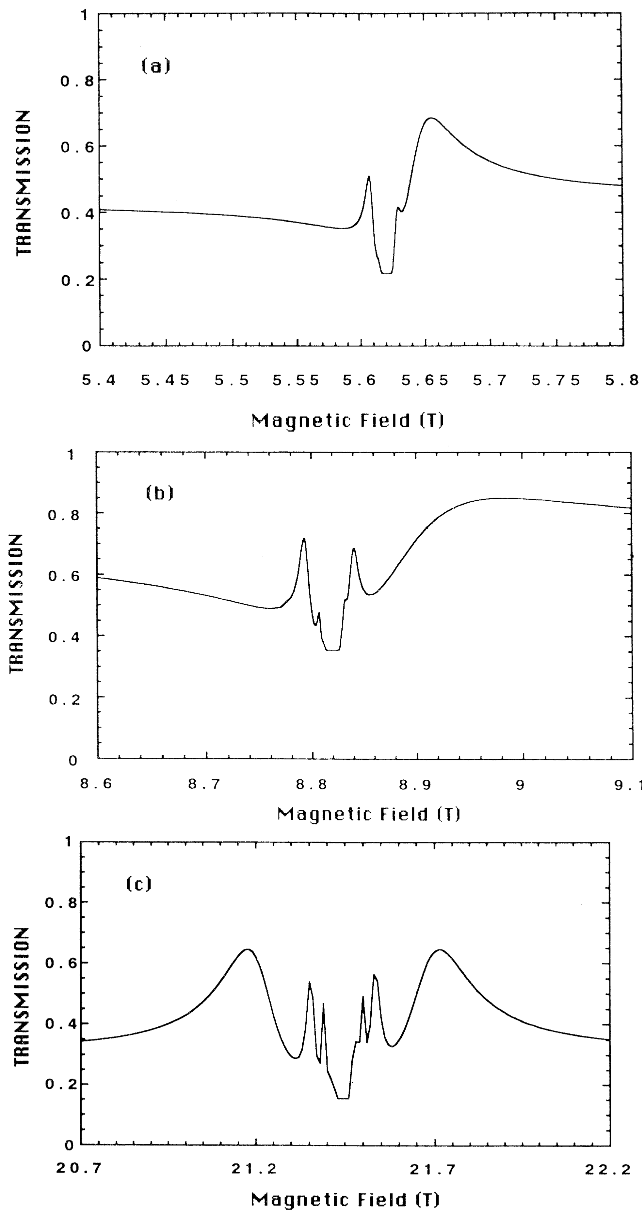

FIG. 3. The calculated transmission curves with interferences for the same three energies in which the experimental data have been collected (see Figs. 1 and 2): (a) $\lambda=1899.9$ μm; (b) $\lambda=1222$ μm; and (c) $\lambda=496$ μm. The following parameters have been used for calculations: $\chi_0=0.001$ (in mks units), $\varepsilon_l=10.8$, $\tau=7.10^{-9}$ s, and thickness $d=0.95$ mm.

intensities of the structures seen in the transmission, one can expect that transitions between pair levels could only be a few times weaker than these due to the absorption in single Mn ions. Therefore, one can expect changes of magnetization of the same order. This, however, is not observed.

In order to better understand our measurements of the transmission and of the magnetization dynamics, we now model the transmission line shapes introducing geometrical interferences (Eq. 4.20 from Ref. 15). To calculate the optical constants (index of refraction n and extinction coefficient k) involved,

$$[n(\omega)+ik(\omega)]^2=\varepsilon_l[1+\chi_r(\omega)+i\chi_i(\omega)] , \quad (1)$$

the following model is used.

The sample has a given dielectric constant ε_l. The complex dynamic susceptibility $[\chi_r(\omega)+i\chi_i(\omega)]$, due to the MD transitions between Mn spin levels, is changing with magnetic field as[16]

$$\chi_r(\omega)=\chi_0\frac{1\pm\omega_0(\omega\pm\omega_0)\tau}{1+(\omega\pm\omega_0)^2\tau^2} ,$$
$$\chi_i(\omega)=\chi_0\frac{\omega\tau}{1+(\omega\pm\omega_0)^2\tau^2} , \quad (2)$$

where χ_0 is the static magnetic susceptibility, ω_0 the resonance frequency, and τ the "relaxation" time responsible for the width of the resonance (usually T_2); $\pm$ signs are related to the σ^+ and σ^- circular polarizations, respectively. Parameters suitable for low composition $Cd_xMn_{1-x}Te$ (approximated after Refs. 12 and 16) were used for numerical calculations. We assume totally unpolarized light, therefore the total transmission T is an average $(T_{\sigma^+}+T_{\sigma^-})/2$.

The results obtained are plotted in Fig. 3. It is clear that interferences can be responsible for the experimentally observed features. It should be noted that only interferences can explain the observed *strong and sharp* rise of the transmission in the region of the absorption minimum. With increasing temperature the absorption becomes weaker and the structure starts to look as a simple absorption line. The data for the powder sample showing a simple line[5] are understood as being free from interferences. The model presented here could partially explain both the observed complicated transmission line shapes and the single lines observed in the dynamic magnetization data.

It was also attempted to study the ESR line shape in the Faraday configuration for different circular polarization. No significant dependence of the absorption on the direction of circular polarization was recognized. Although the degree of polarization was rather low, this result supports the idea that the FIR ESR absorption in $Cd_xMn_{1-x}Te$ exists in both circular polarizations. It suggests that the absorption is not only due to MD transitions, but that also ED transitions can be involved.

IV. CONCLUSION

In summary, ESR line shapes were studied in samples of the diluted magnetic semiconductor $Cd_{1-x}Mn_xTe$ for various Mn compositions at high magnetic fields and at low temperatures. The spin system was excited by using pulsed far-infrared radiation. The longitudinal magnetization dynamics was monitored with a pick-up coil. Fine structure observed in the transmission line shapes was not observed in the coil signal. Therefore, we conclude that these fine structures and the sharp rise in transmission around the resonance are due to the geometrical interferences.

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