# High-field magnetic resonances in $Cd_{1-x}Mn_xTe$ at far-infrared energies

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Magnetic resonances have been studied in $Cd_{1-x}Mn_xTe$ as a function of the concentration $x$, by means of far-infrared magnetotransmission at fields up to $B = 22$ T. For $x \geq 0.3$ an antiferromagneticlike resonance is observed, having a $g$ factor equal to 2. This resonance extrapolates to a finite zero-magnetic-field excitation energy which increases with concentration $x$. The results are explained in terms of single-magnon excitation. For $x \leq 0.1$ EPR of the $Mn^{2+}$ ions is observed, showing a significant line broadening which increases with magnetic field.

## I. INTRODUCTION

Magnetic properties of the semimagnetic semiconductor $Cd_{1-x}Mn_xTe$ at low temperatures have recently been the subject of several experimental and theoretical investigations,[1-13] due to the fact that this system is very well suited for the study of short-range magnetic interactions in diluted materials. As the manganese concentration $x$ increases, the crystal undergoes a spin-glass transition, and above $x \approx 0.6$ a transition to a short-range antiferromagnetic phase.[13] However, the details of the magnetic phase diagram, the magnetic structures in the ordered phase, and the precise critical concentrations $x_c$ for the onset of the different magnetic excitations[10-12] are not fully understood. For instance, the magnetic susceptibility and specific-heat investigations[9] of $Cd_{1-x}Mn_xTe$ at low temperatures have indicated a short-range magnetic ordering already below the critical concentration $x \approx 0.6$, which was later confirmed by neutron scattering experiments.[13] In a subsequent series of Raman scattering investigations[1,14,15] antiferromagneticlike excitations interpreted as magnon modes were also observed at concentrations well below $x = 0.6$. These resonances were attributed to small antiferromagnetic domains in the sample. Two different interpretations of these magnon modes have been proposed, namely, one-magnon excitations at the Brillouin-zone center,[1] or two-magnon processes at the zone boundary.[14] Later results of inelastic neutron scattering are more in agreement with the single magnon model proposed by Ramdas,[1] but its validity is still under dispute.[11,16]

To understand more fully these excitations we present in this report the results of magnetotransmission experiments performed on $Cd_{1-x}Mn_xTe$ samples having a wide range of manganese concentrations. The experiments were carried out in magnetic fields up to 22 T in the far-infrared (FIR) range of the spectrum. Because the applied excitation frequencies are over an order of magnitude higher than in conventional EPR experiments, we can now observe the magnetic resonances at low temperatures over the whole range of manganese concentrations.

A pronounced feature of the EPR spectra in $Cd_{1-x}Mn_xTe$, as well as in other semimagnetic semiconductors,[17] is a significant increase of the resonance linewidth upon decrease of the temperature or increase of the manganese concentration. Typical EPR experiments at low magnetic fields have consequently been limited until now to either low concentrations or high temperatures. This experimental drawback becomes increasingly worse at concentrations in excess of $x \approx 0.2$ due to resonance shifts to lower energies as have been observed recently.[18] Therefore, the use of very high magnetic fields and consequently high excitation energies, as applied in our experiment, are necessary to investigate the low-temperature resonances for higher manganese concentrations $x$.

For large $x$ the resonance energies covered in our experiment correspond to previously observed energies of collective excitations in the ordered (antiferromagnetic) phase, whereas for small $x$ ($< 0.01$) single $Mn^{2+}$ EPR modes can be observed up to the highest magnetic fields. Furthermore, at these high fields at low concentrations practically all $Mn^{2+}$ spins will be lined up because the value of the external magnetic field will approach the value of the internal exchange fields, hence allowing a study of magnetic resonances near complete magnetic saturation.

## II. EXPERIMENT

The single crystals of $Cd_{1-x}Mn_xTe$ with $0.002 \leq x \leq 0.65$ were grown by a modified Bridgeman technique at the Institute of Physics in Warsaw. All samples were cleaved along the (110) plane and polished in the form of disks having diameters from 6 to 10 mm and thicknesses from 1–5 mm.

Magnetotransmission measurements were made in the Faraday configuration at $T = 4.2$ K. At fixed laser frequencies the transmission was measured as a function of the magnetic field. The FIR radiation was generated by an optically pumped FIR molecular laser. A standard lightpipe system was used to focus the radiation onto the

sample. A liquid-helium-cooled detector made of an Allen Bradley carbon resistor was used. A Polyhelix-Bitter solenoid provided dc magnetic fields up to 22 T. For a few samples some measurements at very low excitation energies ($v < 200$ GHz) have been performed using a backward-wave oscillator.

## III. RESULTS AND DISCUSSION

### A. High concentrations

Figure 1 shows the transmission for six $Cd_{1-x}Mn_xTe$ samples with $x$ between 0.002 and 0.6 as a function of magnetic field. The frequency of the incident radiation was 561 GHz. The transmission values were scaled at $B = 0$ T to permit comparison between the different spectra. Note that with increasing manganese concentration the resonance shifts to lower fields and the linewidth increases as observed before.[18,19] However, the low-field measurements were unable to distinguish whether the observed shift to lower fields with concentration was caused by changes in the $g$ factor[19] or by the presence of internal fields.[20] In Fig. 2, therefore, the positions of the resonance energies are plotted versus magnetic field for several concentrations. The solid line marks a $g = 2$ EPR transition and it is clearly shown that all dashed lines, which present least-squares fit to the measurements, have practically the same slope as the $g = 2$ resonance. This figure also shows that for $x \geq 0.3$ a linear extrapolation of the resonance frequency to zero magnetic field yields a finite resonance energy which can be attributed to an antiferromagnetic resonance mode (AFMR).

Although linear extrapolation from fairly high fields to zero magnetic fields may introduce errors,[21] it is still interesting to compare these results with those of other low- or zero-field studies of the magnetic excitations in Mn-
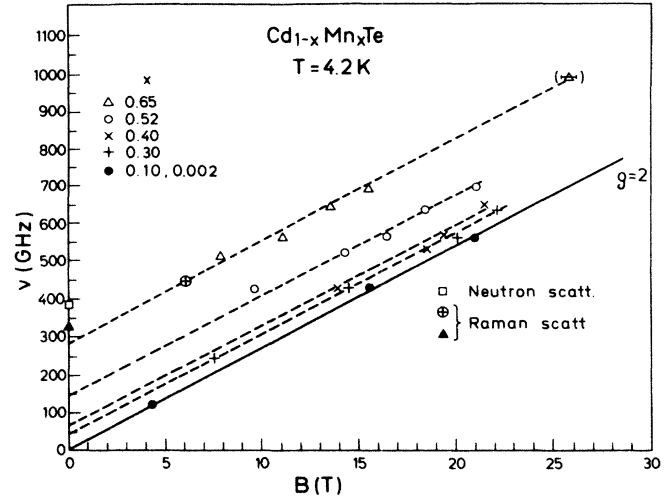


FIG. 2. The resonance frequency versus the magnetic field for several manganese concentrations. The position of the high-field resonance at 989 GHz for $x = 0.65$ presents an extrapolated value, because only a part of the resonance could be observed below $B = 22$ T. The solid line marks a $g = 2$ EPR transition. The dashed lines present a least-squares fit to the experimental results. For comparison the magnetic excitation energies obtained from neutron scattering ($\square$, Refs. 13 and 16) and from Raman scattering ($\oplus$, Ref. 1 and $\blacktriangle$, Ref. 14), are also indicated.

rich $Cd_{1-x}Mn_xTe$. For this purpose we have included also in Fig. 2 some results of nonelastic neutron scattering[16] (NS) and Raman scattering[1,14] (RS) experiments. Corresponding points in the figure mark the positions of the resonances observed by these techniques. The agreement with the RS data at $B = 6$ T is especially good. The deviations for the zero-field RS and NS data are possibly due to slight differences in chemical composition of the samples used. Moreover those measurements were done at either lower (1.8 K) or at higher (10 K) temperatures which induces some changes in the resonance energies. To display better the zero-field resonances, their energies are plotted in Fig. 3 as a function of the manganese concentration $x$ together with the RS and NS results.[1,14,16] This figure suggests that the low-temperature far-infrared resonance as observed in our experiment for $x \geq 0.3$ is an AFMR at finite magnetic fields. Moreover, for $x = 0.65$ the resonance frequency agrees well with the magnon excitation observed in neutron scattering experiments. With decreasing concentration $x$ this resonance energy decreases and finally disappears near $x \approx 0.25$, in reasonable agreement with the value of the percolation limit $x_c = 0.19$ for a fcc lattice. Such behavior of AFMR has been observed before in diluted antiferromagnets.[22] Furthermore, the present observations agree well with the Raman results[1] which have shown the persistence of a magnon mode down to quite low-$x$ values.

Now we would like to address the problem of the origin of the magnetic Raman excitation as observed by Ramdas,[1] and Grynberg and Picquart.[14] Figures 2 and 3 strongly indicate that the excitation energies as observed in these experiments correspond to the resonances seen in
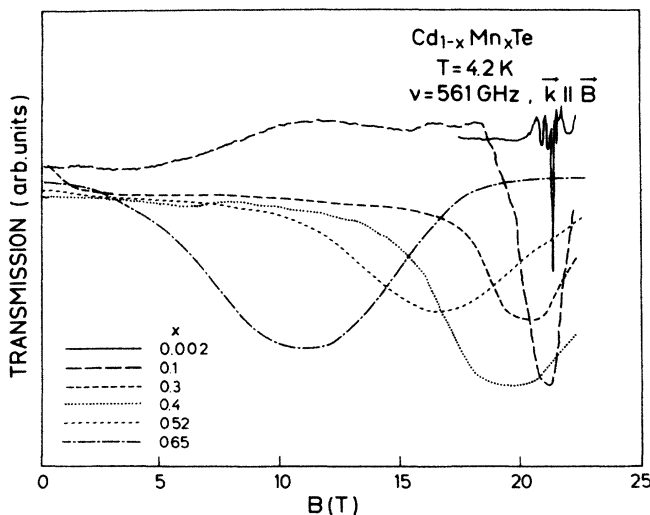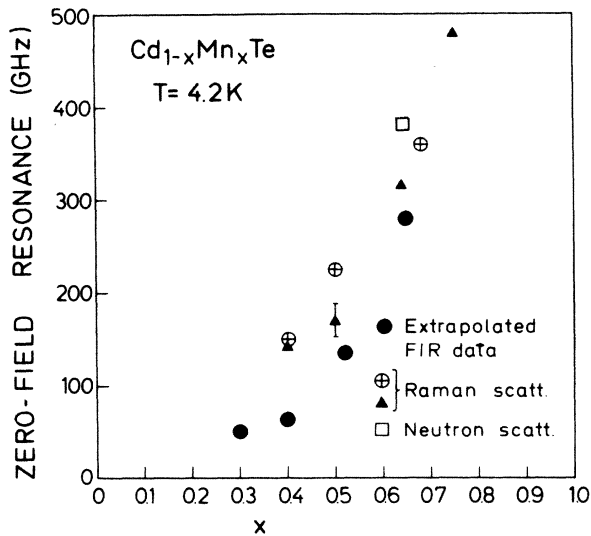


FIG. 1. The transmission of 561 GHz radiation as a function of magnetic field B for different manganese concentrations. The transmission values are relatively scaled to allow easy comparison.

FIG. 3. The zero-field resonance frequency as a function of the manganese concentration $x$. $\oplus$, Raman results from Ref. 1; $\blacktriangle$, Raman results from Ref. 14; and $\square$, neutron scattering result from Ref. 16. $\bullet$ represent linear extrapolated points of FIR data from Fig. 2.

the magnetotransmission spectra, and as such are likely to have the same physical origin. Ramdas[1] describes the magnetic Raman peak as a single quantum (magnon) excitation in the antiferromagnetic ordered phase of $Cd_{1-x}Mn_xTe$. On the other hand, Grynberg[14] analyzes the results as a two-magnon effect, excluding single magnon excitation on grounds of a symmetry argument and the absence of higher-energy magnetic excitations. As pointed out by Sievers,[23] in a two-magnon process one magnon is emitted from each of the normally degenerated spin-wave branches at the magnetic Brillouin-zone boundary. An external magnetic field splits these branches, of one magnon the energy is decreased, and of the other one it is increased by the same amount. Therefore such a two-magnon excitation is expected to have a resonance energy independent of the magnetic field. As Fig. 2 clearly shows, the resonance frequency of the excitation has a dependence on magnetic field corresponding to a $g$ factor equal to 2. We must conclude therefore, that the observed antiferromagnetic mode will be due to a single magnon excitation, and not to a two-magnon process.

### B. Low concentrations

Below $x \approx 0.1$ the EPR resonance position, as seen in Fig. 1 is very well described by $g = 2$ without a finite zero-field resonance energy, indicating the absence of antiferromagnetic ordering. The resonance positions versus magnetic field have become rather independent of the concentration. Therefore, in principle, we observed now the resonance energy of single $Mn^{2+}$ spins, but having a resonance linewidth affected by both dipole-dipole, and exchange interaction. The first mechanism tends to broaden the linewidth for concentrations $x > 0.01$, whereas the latter, due to nearest neighbor interaction for $x > 0.03$ will narrow the observed resonance (exchange narrowing). Because of this exchange narrowing, usual EPR
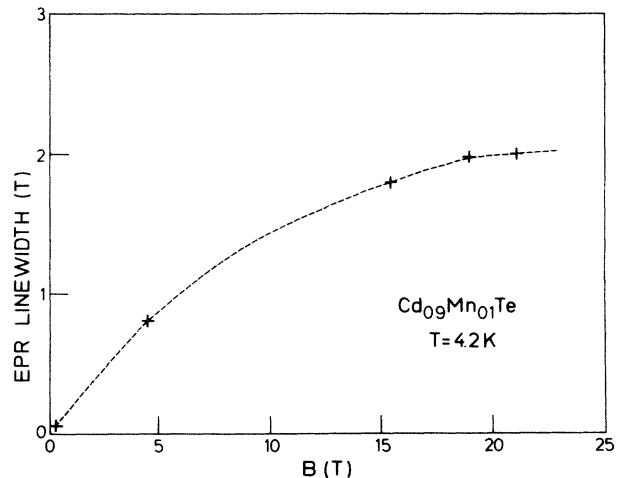


FIG. 4. The linewidth of the EPR signal of the $x = 0.1$ sample as a function of magnetic field, at $T = 4.2$ K. The dotted line presents only a guide to the eye.

measurements at low magnetic fields yield a linewidth for $x = 0.1$ at $T = 4.2$ K of approximately, 400 G. As shown in Fig. 1 however, the linewidth of the $x = 0.1$ resonance is much larger. This can be due to the very strong applied magnetic field, which is of the same order of magnitude as the internal fields, hence reducing the importance of the exchange interaction. Measured linewidths at a few magnetic field values for $x = 0.1$ are given in Fig. 4, and show an increase with magnetic field which tends to saturate above $B \approx 18$ T. These observations support the idea of the external magnetic field approaching the strength of the internal exchange fields. Figure 5 shows a fit to the line shape using a simple Bloch two-level spin model, for noninteracting localized spins, taking into account the effects of electromagnetic wave propagation in a parallel slab.[24] The line shape was fitted with a 0.75 Lorenzian,
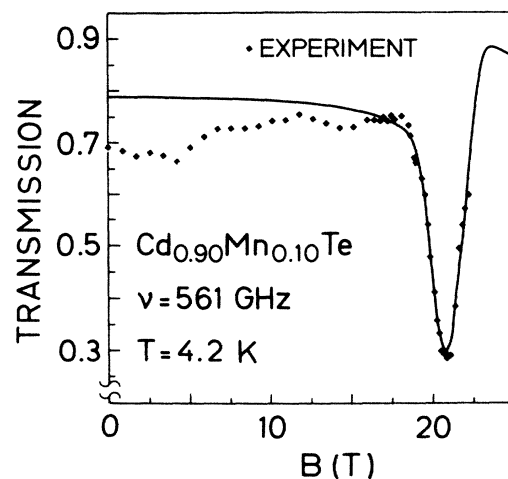


FIG. 5. The transmission (arbitrary units) of 561 GHz radiation as a function of magnetic field for the $x = 0.1$ sample, together with a theoretical fit, using a model for separated spins as described in the text.

0.25 Gaussian function. The static sample magnetization was used as a fitting parameter and found to be equal to $7 \pm 1$ emu/g at $B = 21$ T, in agreement with results of high-field magnetization measurements which gave 6.1 emu/g at the same field.[25]

In summary, we have measured magnetic resonances at high magnetic fields in $Cd_{1-x}Mn_xTe$ for a broad range of concentrations $x$. At low concentrations a line-broadened paramagnetic resonance is observed having a $g$ factor equal to 2. At concentrations $x \geq 0.3$ an antiferromagnetic resonance is observed which complies well with previously observed magnon modes using Raman and neutron scattering techniques. On grounds of the observed

magnetic-field dependence of this excitation, the resonance is ascribed to a single-magnon mode having a $g$ factor equal to 2, and not to a two-magnon mode, as has been suggested recently.

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