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RAMAN SCATTERING BY PHONONS AND MAGNONS IN MBE-GROWN $Cd_{1-x}Mn_xTe$ LAYERS*

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Collective excitations have been analyzed for $Cd_{1-x}Mn_xTe$ epilayers ($0.66 \le x \le 1$) by the Raman scattering studies performed at temperatures from 7 K to 295 K. Apart from the lattice optical modes magnetic excitations (magnons) were observed at sufficiently low temperatures.

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Bulk zinc-blende crystals of $Cd_{1-x}Mn_xTe$ can be obtained by equilibrium growth techniques only for $x < 0.7$. For higher Mn contents the system crystallizes in hexagonal NiAs structure. However, one can obtain $Cd_{1-x}Mn_xTe$ for $0.7 \le x \le 1.0$ in a metastable form with the zinc-blende structure by MBE on substrates of the cubic symmetry. In this paper thin films of zinc-blende $Cd_{1-x}Mn_xTe$ with $0.7 \le x \le 1.0$ were grown using an EPI 620 MBE system on semi-insulating (001)GaAs wafers. A thin (100–1000 Å) ZnTe layer was employed in order to reduce the strong mismatch between a typical 2 μm thick CdTe buffer layer and GaAs substrate and to stabilize the growth in the (100) direction. A mixed crystal composition was determined by X-ray diffraction methods.

Raman scattering experiments were performed in a quasi backscattering geometry. Spectra were recorded using a double monochromator equipped with holographic grating and a S20 photomultiplier or CCD detecting system. For the excitation Ar^+ laser lines (457.9 nm, 476.5 nm and 514.5 nm) and Kr^+ lines (647.1 nm and 530.9 nm) were applied.

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Raman spectra taken for the mixed crystal exhibit two pairs of sharp lines characteristic for the zone center "MnTe-like" and "CdTe-like" LO-TO modes. LO- and TO-frequencies obtained as a function of Mn concentration well correspond to the previously reported values received for the bulk crystals (see, e.g. [1]). LO and TO phonon frequencies for cubic MnTe directly determined at helium temperature are equal to (218 ± 1) cm^{-1} and (190 ± 1) cm^{-1}, respectively. These values are in perfect agreement with the values recently determined by other methods [2, 3].

For compositions of the mixed crystals close to MnTe an additional structure in the Raman spectra corresponding to the Te precipitations is found [4] (Fig. 1a). For a few MnTe samples these findings are also confirmed by X-ray diffraction measurements (for details see [5]).

The most exciting effect observed in our samples was spin-wave excitation (magnon) found at low temperatures (see Fig. 1a) for all samples except the epilayer with the lowest composition ($x = 0.66$). For this particular sample in the spectral region corresponding to the expected magnon peak frequency an efficient Rayleigh light scattering appeared. Magnons were seen in the bulk $Cd_{1-x}Mn_xTe$ samples with $x \leq 0.7$ more than 10 years ago [1, 6], but have not been reported till now for a thin layer of any semimagnetic semiconductor grown by the MBE technique. Variation of the magnon peak frequency with crystal composition is shown in Fig. 1b. The magnon peak frequency for zinc-blende MnTe is as high as (34.0 ± 0.5) cm^{-1} and within the experimental error is the same for all investigated MnTe samples. As one can see the present data substantially complete the results determined in the past for the bulk crystals. For a few samples the temperature dependence of the magnon frequency was determined. From such measurements a temperature of the magnetic phase transition can be estimated. As it has been mentioned in Ref. [1] assuming that the magnetic ordering for high values of x

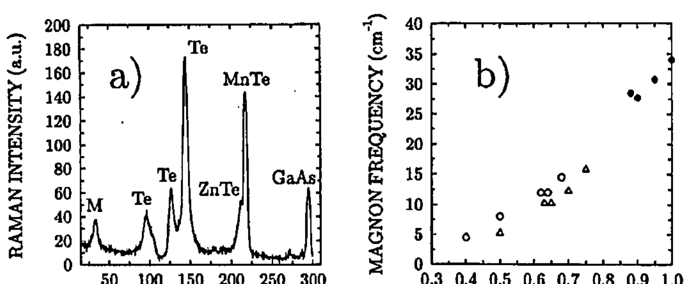

Fig. 1. (a) Part of the Raman spectrum taken at 17 K for one of investigated MnTe samples (thickness of MnTe slab — 2.3 μm, thickness of ZnTe buffer layer — 1000 Å, Ar^+ laser excitation line — 476.5 nm). The letter M denotes the structure due to the Raman scattering on magnons. (b) Dependence of the magnon peak frequency on Mn concentration in the mixed crystal. Triangles: data for the bulk crystals taken from Ref. [1], circles: data for the bulk crystals taken from Ref. [6], full circles: our results.

corresponds at low temperatures to antiferromagnetic phase, one can calculate the temperature dependence of the magnon peak frequency. For this purpose it is necessary to find the temperature dependence of the magnetization corresponding to the analyzed mixed crystal composition. Using the same model as that presented in Ref. [1] it was estimated that the exchange integral describing the interactions between the nearest-neighbors $J_{NN} = -(5.4 \pm 0.3)$ K. This value is in reasonable agreement with the predictions and estimations based on other kinds of investigations.

The magnetic phase diagram for $Cd_{1-x}Mn_xTe$ semimagnetic semiconductor is intensively investigated and verified recently by different teams using a SQUID magnetometer for the magnetization measurements. Such diagram originally determined from the specific-heat and magnetic-susceptibility studies suggested that the paramagnetic spin-glass transition occurs for the composition range $0.17 < x < 0.6$ [7]. For a higher Mn content the paramagnetic–antiferromagnetic phase transition should appear. The magnetic phase diagram of $Cd_{1-x}Mn_xTe$ [8–10] shows that the results obtained for the bulk crystals and epilayers are in very good agreement for the mixed crystal composition $x < 0.6$. It seems that for a higher Mn content in the slab (and, in particular, in the composition range corresponding to the bulk hexagonal crystal structure) the different magnetic behavior is observed. Moreover, the magnetic transition temperature of epilayers is noticeably higher than that of the bulk crystals (compared for the same high composition value). More systematic Raman scattering measurements could help one to verify this discrepancy and to check the possible influence of the internal stress on the reported effect.

According to the literature data the magnetic phase transition temperature determined using SQUID is slightly below 70 K for cubic MnTe [8–10]. This value confirms the result of previous neutron scattering experiments [11] and is in agreement with our data. Nevertheless, there still exists the open problem concerning the character of the magnetic phase ordering for the mixed crystals with high enough composition values. Magnons can be generated both in the spin-glass phase and in the antiferromagnetic phase, therefore it is not possible to distinguish between these two possibilities only on the basis of the Raman scattering data.

In conclusion we would like to stress that the successful observation of the magnetic excitations in the thin $Cd_{1-x}Mn_xTe$ epilayers opens the real possibility of the Raman scattering investigations of the magnetic excitations in quantum structures and superlattices. It is also a direct proof of the very high quality of the samples.

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References

[1] S. Venugopalan, A. Petrou, R.R. Gałązka, A.K. Ramdas, *Solid State Commun.* **38**, 365 (1981); S. Venugopalan, A. Petrou, R.R. Gałązka, A.K. Ramdas, S. Rodriguez, *Phys. Rev. B* **25**, 2681 (1982).

[2] M. Dean Sciacca, A.J. Mayur, Eunsoon Oh, A.K. Ramdas, S. Rodriguez, *Solid State Commun.* **88**, 711 (1993).

[3] K. Ando, K. Takahashi, Y. Takeuchi, *Mat. Res. Soc. Proc.* **279**, 855 (1993).

[4] A.S. Pine, G. Dresselhaus, *Phys. Rev. B* **4**, 356 (1971); W. Richter, *J. Phys. Chem. Solids* **33**, 2123 (1972).

[5] W. Szuszkiewicz, M. Jouanne, E. Dynowska, E. Janik, G. Karczewski, T. Wojtowicz, J. Kossut, W. Gębicki, in: *Proc. Int. Conf. on Semconductor Heteroepitaxy, Montpellier 1995* (in press).

[6] M. Grynberg, M. Picquart, *J. Phys. C, Solid State Phys.* **14**, 4677 (1981); M. Picquart, M. Grynberg, *J. Appl. Phys.* **53**, 8166 (1982).

[7] R.R. Gałązka, S. Nagata, P.H. Keesom, *Phys. Rev. B* **22**, 3344 (1980).

[8] K. Ando, K. Takahasi, T. Okuda, *J. Magn. Magn. Mater.* **104–107**, 993 (1992); K. Ando, H. Akinaga, *J. Magn. Magn. Mater.* **140–144**, 2029 (1995).

[9] M. Sawicki, S. Koleśnik, T. Wojtowicz, G. Karczewski, E. Janik, M. Kutrowski, A. Zakrzewski, E. Dynowska, T. Dietl, J. Kossut, *Acta Phys. Pol. A* **87**, 169 (1995).

[10] J. Pietruczanis, W. Mac, A. Twardowski, G. Karczewski, A. Zakrzewski, E. Janik, T. Wojtowicz, J. Kossut, in: *Proc. European Workshop on II–VI Semiconductors and Int. Workshop on Semimagnetic (Diluted Magnetic) Semiconductors*, Eds. H. Heinrich, J.B. Mullin, Trans Tech Publ., *Materials Science Forum* **182–184**, 687 (1995).

[11] P. Kłosowski, T.M. Giebułtowicz, J.J. Rhyne, N. Samarth, H. Luo, J.K. Furdyna, *J. Appl. Phys.* **70**, 6221 (1991); T.M. Giebułtowicz, P. Kłosowski, N. Samarth, H. Luo, J. K. Furdyna, J.J. Rhyne, *Phys. Rev. B* **48**, 12817 (1993).