Resonance Raman Scattering by LO Phonons in CdTe/CdMnTe superlattices

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We have observed confined and interface phonons in [111] CdTe/CdMnTe superlattices. With decreasing barrier thickness, a small red shift and an increasing broadening of the Raman resonance with the first heavy-hole exciton has been measured. A dependence of the phonon frequencies on laser energy is discussed in terms of strain present in the samples.

Superlattices (SL's) of diluted magnetic semiconductors offer new possibilities to tune magnetic interactions and to study the effects of dimensionality on magnetic behavior [1]. The aim of this work is the study of the formation of SL's in the CdTe/CdMnTe system by decreasing the thickness between adjacent wells. Resonance Raman scattering is a powerful technique for these studies since it proves electronic excitations [2], which should reflect the changes in dimensionality, it is very sensitive and it is free from extrinsic effects due to impurities and imperfections, present in other optical techniques such as photoluminescence.

The samples were grown by molecular beam epitaxy on (100)–GaAs substrates. A 0.15μm (111)-oriented CdTe buffer layer was followed by a CdTe/$Cd_{1-x}Mn_xTe$ superlattice. Two series of samples with x=0.21 and 0.10, constant well width (d_1=86A) and three different barrier thicknesses, d_2= 86A,40A,20A, with 25,50 and 100 periods, respectively, were investigated; here we will focus on the series with the larger Mn concentration (samples I,II and III, respectively). Raman spectra at 10K were excited with an LD700 cw-dye laser pumped with a Kr^+-laser. The scattered light was analyzed with a double monochromator and detected by photon counting.

The phonon spectra of CdMnTe bulk crystals [3] and CdTe/CdMnTe SL's [4–6] have been studied previously in the literature. Unpolarized Raman spectra in the vicinity of the first heavy-hole exciton (h_1) are shown in Fig.1 for sample I (similar spectra were obtained in parallel and crossed polarizations). Far above from resonance (1.916eV) three structures at 166cm^{-1} (LO_1 CdTe-like), 171cm^{-1} (LO CdTe) and 198cm^{-1}(LO_2 MnTe-like), are resolved in the spectrum. Due to its resonance behavior (see below), we assign the shoulder at $195cm^{-1}$ to a MnTe-like interface mode (IF) [4,7]. As the laser energy approaches h_1 (1.635eV), the LO_2 mode is strongly reduced, while LO and IF are resonantly enhanced. The LO_1 mode, which can propagate in both layers [4], is observed as a shoulder at the low-energy side of LO. At energies below h_1 only the LO–CdTe phonon is seen in the spectra.

The resonance of the LO–CdTe phonon for the three SL's, together with that of the IF-mode for sample I, are shown in Fig.2. The enhancements around $13100cm^{-1}$ and $13300cm^{-1}$ are due to the incoming and outgoing resonances with h_1, respectively [5,6]. We assign the asymmetry on the high-energy side of the resonance to the weakly confined light-hole exciton, whereas the shoulder at $\sim 13900cm^{-1}$ may correspond to the second heavy-hole exciton. The similar behavior of curves a) and b) in Fig.2 clearly indicates the interface character of the phonon labelled IF [7]. The shift of $\sim 25cm^{-1}$ between the outgoing resonances of IF and LO agrees with the energy difference of their frequencies [3,8].

As the barrier width is reduced from 86A to 20A (curves b) and d) in Fig.2) a small, but measurable, red shift, together with an increasing broadening of

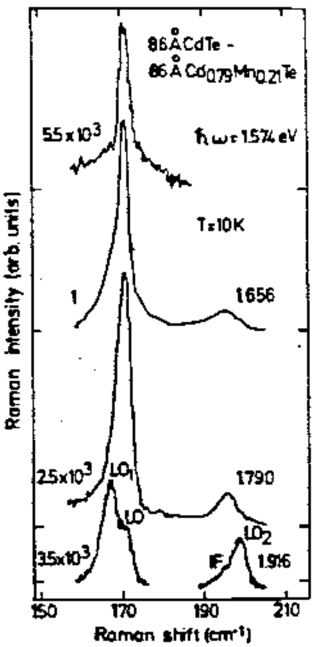

Figure 1: Raman spectra for sample I at 10K for different laser energies.

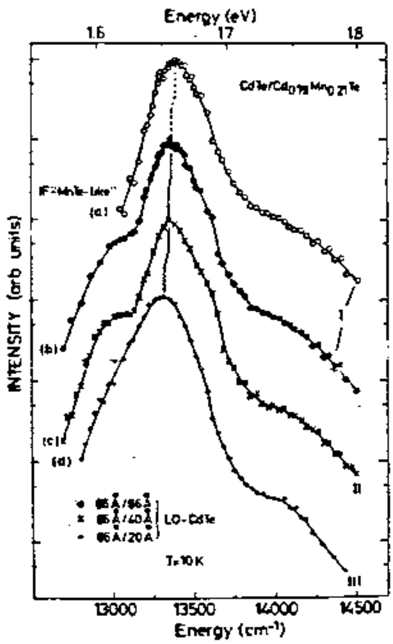

Figure 2: Resonance Raman intensities of the IF- and LO-phonons. The lines are to guide the eye.

the LO-phonon resonance is observed. Taking sample I as a reference, a crude estimation using a Kronig-Penney model with the same parameters as in Ref.6 and a conduction- to valence-band offset ratio of 14:1 [9] obtains an energy shift, ΔE, (broadening, $\Delta\Gamma$) of $15cm^{-1}$($30cm^{-1}$) and $55cm^{-1}$($100cm^{-1}$) for samples II and III, respectively. The measured values are: ΔE=10(±10)cm^{-1}, 40(±15)cm^{-1} and $\Delta\Gamma$=10(±10)cm^{-1}, 70(±10)cm^{-1} for samples II and III, respectively. Both findings indicate the formation of minibands as the barrier width is reduced.

Marked differences between [100] and [111] CdTe/CdMnTe SL's, attributed to the quality of the interfaces and to the presence of strain, have been reported in the literature [4,10]. In particular, a dependence of the phonon frequencies on laser energies in [111] samples has been interpreted on the basis of the dispersion relation for IF modes [4]. We have found a similar dependence on the first runs of our experiments. Figure 3 depicts this dependence for samples I and III. However, in more recent measurements, after cooling the samples several times, we do not find any appreciable shift with laser energy (within ±1cm^{-1}). Our first data cannot be explained using the arguments of Ref.4, and a convincing explanation for the observed dependence is still lacking. Recent transmission electron microscopy [11] and photoluminescence [12] measurements indicate that biaxial compressive strains are present in [100] CdTe films, thinner than 1μm, grown on GaAs. For [111] samples the strain should be smaller than for the [100] case [13]. However, since our samples are below or only slightly above this thickness, it is possible that strain present in the samples has relaxed after the cooling cycles (GaAs and CdTe have a quite different linear expansion coefficient, $6.9\times10^{-6}K^{-1}$ and $4.8\times10^{-6}K^{-1}$, respectively [14]). Figure 4 compares the resonant profile of the LO-phonon in sample III in the first (A) and more recent experiments (B). The shift towards higher energies

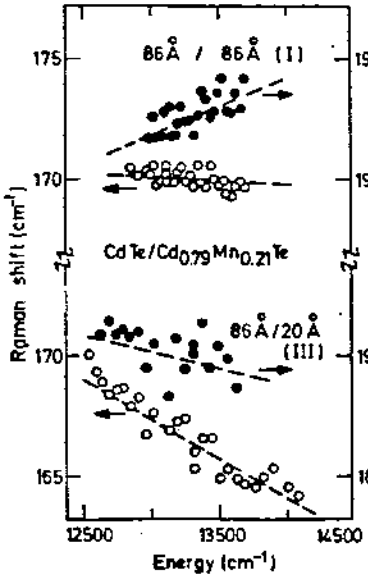

Figure 3: Frequency dependence of Raman phonons as a function of laser energy for samples I and III.

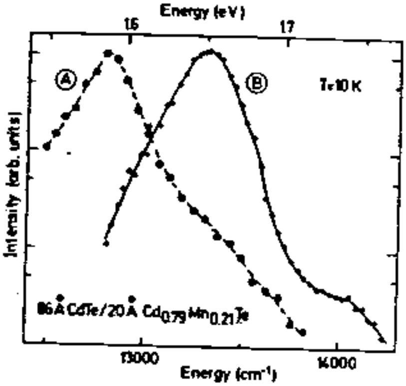

Figure 4: Resonance Raman intensities of the LO-phonon in sample III in the first runs of experiments (A) and after several cooling cycles (B).

suggests a relaxation of the strain in the samples [12].

In summary, we have shown that confined and interface phonons can be observed in {111} CdTe/CdMnTe superlattices. A red shift and an increasing broadening of the LO-phonon resonance with decreasing barrier thickness indicate the formation of minibands in the superlattices.

References

1. D.D. Awschalom, G. Grinstein, J. Yoshino, H. Munekata, and L.L. Chang, Surf. Sci. 196, 649 (1988).
2. M. Cardona in *Light Scattering in Solids*, Topics in Applied Physics, Vol.50, ed. by M. Cardona and G. Güntherodt (Springer-Verlag, New York 1982).
3. S. Venogupalan, A. Petrou, R.R. Galazka, A.K. Ramdas, and S. Rodriguez, Phys. Rev. B 25, 2681 (1982).
4. E.-K. Suh, D.U. Bartholomew, A.K. Ramdas, S. Rodriguez, S. Venogupalan, L.A. Kolodziejski, and R.L. Gunshor, Phys. Rev. B 36, 4316 (1987)
5. S.-K. Chang, H. Nakata, A.V. Nurmikko, L.A. Kolodziejski, and R.L. Gunshor, Journal de Physique C5, 345 (1987).
6. L. Viña, L.L. Chang, and J. Yoshino, Journal de Physique C5, 317 (1987).
7. A.K. Sood, J. Menendez, M. Cardona, and K. Ploog. Phys. Rev. Lett. 54, 2115 (1985).
8. J. Menendez and M. Cardona, Phys. Rev. B 31, 3969 (1985).
9. S.-K. Chang, A.V. Nurmikka, J.-W. Wu, L.A. Kolodziejski, and R.L. Gunshor, Journal de Physique C5, 367 (1987).
10. S.-K. Chang, A.V. Nurmikko, L.A. Kolodziejski, and R.L. Gunshor, Phys. Rev. B 33, 2589 (1987).
11. J. Petruzzello, D. Olego, S.K. Ghandhi, N.R. Taskar, and I.B. Bhat, Appl. Phys. Lett. 50, 1423 (1987).
12. D. J. Olego, J. Petruzzello, S.K. Ghandhi, N.R. Taskar, and I.B. Bhat, Appl. Phys. Lett. 51, 127 (1987).
13. N. Otsuka, L.A. Kolodziejski, R.L. Gunshor, S. Datta, R.N. Bicknell, and J.F. Schetzina, Appl. Phys. Lett. 46, 860 (1985).
14. Landolt-Börnstein Tables, Vol.17, ed. by O. Madelung, M. Schulz and H. Weiss (Springer-Verlag, New York, 1982).