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RAMAN SCATTERING AND EXCITATION SPECTROSCOPY IN CdTe/CdMnTe SUPERLATTICES

L. VIÑA[1], L.L. CHANG and J. YOSHINO[2]

IBM Thomas J. Watson Research Center, P.O. Box 218, Yorktown Heights, NY 10598, U.S.A.

Abstract.- We have observed oscillatory structure in the excitation spectra of CdTe/$Cd_xMn_{1-x}Te$ superlattices. A comparison of these spectra with conventional Raman spectra shows that the structures correspond to first and higher order LO-phonons of the CdTe wells and the CdTe/$Cd_xMn_{1-x}Te$ barriers, as well as combination of them. A strong enhancement in Resonance Raman scattering of both the CdTe and the CdMnTe phonons, at the energy of the heavy-hole exciton of the superlattice, suggests a small valence-band offset between the two materials.

INTRODUCTION

High quality superlattices (SL's) based on $Cd_xMn_{1-x}Te$ have been grown in the last years using epitaxial techniques[1-3]. Although the optical properties of these SL's have been profusely investigated with methods such as photoluminescence (PL) and excitation spectroscopy (PLE)[4], Raman studies are scarce[5-7]. The phonon spectra of CdTe and $Cd_xMn_{1-x}Te$ have been studied in great detail in the literature[8], and can be used as a reference to compare new results in the SL's. Here we present a combined Resonance Raman scattering and PLE study of CdTe/CdMnTe SL's. A correlation between the Raman spectra and striking oscillatory structure in PLE is established. On the other hand, the resonant behavior of the CdTe and $Cd_xMn_{1-x}Te$ phonons indicates a small valence-band offset between the two materials.

EXPERIMENTAL RESULTS AND DISCUSSION

The samples used in our study have been grown by molecular beam epitaxy on (100)-oriented GaAs substrates. A thick (~200nm) (111)-CdTe buffer layer was followed by a CdTe/$Cd_{0.87}Mn_{0.13}Te$ superlattice. Three different samples with constant well-width L_z =100A and barrier-widths L_b=100,50 and 25A were investigated. Details of the growth technique and characterization have been described in a previous publication[3].

The samples were mounted on a variable temperature cryostat and kept in a He-gas atmosphere at 4.8K. An LD700 dye, pumped by a Kr^+-ion laser, was used to excite the samples with power densities below 1W/cm² and 5W/cm² for the PLE and the Raman experiments, respectively. The scattered light was analyzed with a 3/4m-Spex double monochromator and detected with a GaAs-cathode photomultiplier.

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(1) Permanent address : Instituto de Ciencia de Materiales, Zaragoza, Spain
(2) Tokyo Institute of Technology, Tokyo, Japan

Figure 1 depicts the PL (dashed line) and the PLE (solid line) spectra of a 25-periods superlattice with 100A-wide wells and barriers. A Stokes shift of 8meV between the PL and the PLE, which in GaAs/GaAlAs SL's has been attributed to well-width fluctuations[9] or to impurities[10], is clearly observed in the figure. The PLE spectrum shows three broad structures (1.636, 1.653 and 1.695eV) together with sharp peaks, which correspond to first and higher order LO-CdTe phonons. The vertical-downward arrows show the results of a calculation, in the envelope-function approximation, for the first heavy- (h_1) and light-hole (l_1) excitons, as well as for excitons associated with the second conduction- and valence-band levels [h_2, h_{12}, h_{21}, where the first (second) subindex indicates conduction- (valence-) band level]. Effective masses[11] m_e^*=0.096, m_h^*=0.4 and m_l^*=0.1, a free-exciton dependence on Mn concentration[12], x, E(x)=1.5976+1.564x(eV), and a somehow arbitrarily chosen 90-10% rule for the conduction- and valence-band discontinuities were used for the calculation. Taking into account the uncertainties in the parameters used for the calculation and strain effects in these lattice-mismatched superlattices[12], the agreement with the experiment is satisfactory and hints to a small valence-band offset between the two materials of the SL.

Excitation spectra of a SL with 50A-wide barriers are shown in Fig.2. As the detection energy (arrow) is moved toward higher values, sharp periodic structures are resolved. These dominate completely the spectrum obtained at the high-energy tail of the PL (dashed line). The energy distance of the first two peaks to the detection energy is 21.3meV (LO) and 24.3meV (LO_2), which corresponds to the CdTe (well) and MnTe-like (in CdMnTe) LO-phonons[8]. The additional structures, with a period between the main peaks of 21.3meV, correspond to higher order LO phonons of the CdTe wells and the CdTe/$Cd_{0.87}Mn_{0.13}$ barriers and combinations of them. Oscillatory structure in the PLE spectra of bulk CdTe has been observed previously [13]. Since phonon lines can be observed below and above the h_1 exciton, and a second periodicity, slightly higher than the LO frequency, is absent in our spectra, hot-exciton formation and hot-electron effects can be ruled out, leaving Resonance Raman scattering as the only mechanism responsible for the observed periodic structure[13]. Notice that the phonon peaks are also observed in the PL spectrum, however a much higher sensitivity is obtained in the PLE spectra.

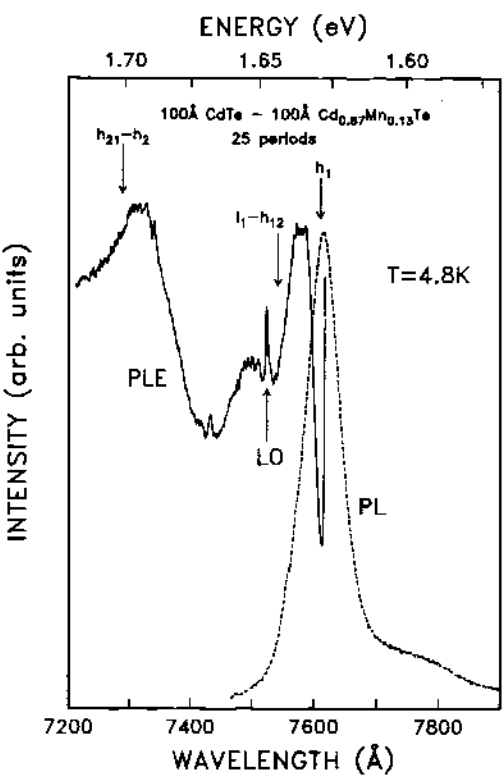

Figure 1: Photoluminescence (PL) and excitation spectra (PLE) of a CdTe/CdMnTe superlattice with 100A-wide wells.

Important information about electronic excitations in a semiconductor can be obtained from the resonant behavior of the Raman intensities, which occur whenever the energies of the incident (incoming channel) or scattered photons (outgoing channel) coincide with the energy of the excitation[14]. We have investigated first- and higher-orders Resonant Raman scattering in these SL's. Figure 3 shows the results for the first-order scattering at 4.8K for the SL with 100A-wide barriers. The spectra were recorded in the z'(x'x')z', where x' is in the plane of the layers and z' along the SL axis (the energy of the incident photon is indicated for each spectrum). Two well-defined peaks at 172cm^{-1}(LO-CdTe) and 196cm^{-1}(LO_2 MnTe-like), together with a small shoulder at 168cm^{-1}(LO_1 CdTe-like)

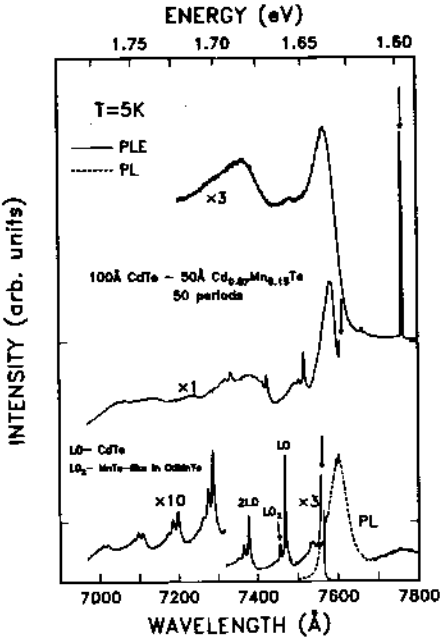

Figure 2: PL (dashed line) and PLE spectra taken at different energies (arrows) of a SL with 50A-wide wells.

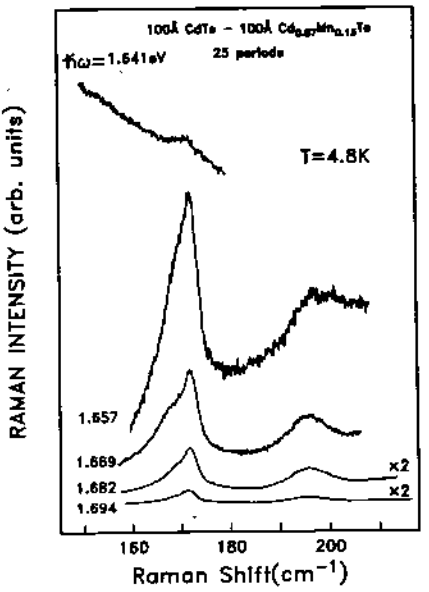

Figure 3: Raman spectra for the same sample as in Fig.1 for different laser energies.

are resolved in a broad background due to the luminescence emission of the sample. The frequencies are in good agreement with the results of Ref. 8 for a Mn concentration x=0.13. As the energy of the scattered photons approaches h_1 (1.636eV), a strong enhancement of the LO and LO_2 phonons is observed.

The intensity of the LO phonon, uncorrected for the absorption in the sample, is plotted in Fig. 4 as a function of laser wavelength. The PLE spectrum is also shown in the figure for comparison. The Raman points have been shifted by 1LO=21.3meV towards lower energies, so that the resonance of the outgoing channel can be directly compared with the h_1 exciton seen in the PLE spectrum. A strong enhancement by ~2 orders of magnitude is observed at the energy of h_1. A second structure at 1.615eV, 1LO phonon frequency below h_1, corresponding to the incoming channel is also observed. The asymmetry in the lineshape of the outgoing resonance, as well as its stronger enhancement, have been attributed to impurity effects[15]. A much weaker enhancement for the l_1 exciton can be observed in the figure; in this case the incoming and outgoing channels show a comparable behavior, in agreement with recent results of Chang et al.[6]

An enhancement of the LO_2 phonon at the energy of h_1 has also been observed, although in this case the resonance could not be traced below the PLE peak due to the luminescence emission of the sample and to the lower intensity of LO_2 compared to that of LO. In the energy range of 1.75-1.80eV, corresponding to the band gap of CdTe/$Cd_{0.87}Mn_{0.13}Te$, the phonons of the barriers and the wells are again resonantly enhanced. These two facts indicate a large penetration of the hole wavefunctions into the barrier material, and therefore a a small valence-band offset between the two materials. Our results contradict recent Raman studies of Suh et al.[7], who have found a selective resonance enhancement of the LO phonons in the barriers and in the wells. The difference could probably lie in the larger Mn concentration used in their samples (x>0.25). Furthermore, we have not observed any frequency dependence on the incident photon energy of the LO_1 and LO_2 phonons, reported for [111] CdTe/CdMnTe SL's in Ref. 7.

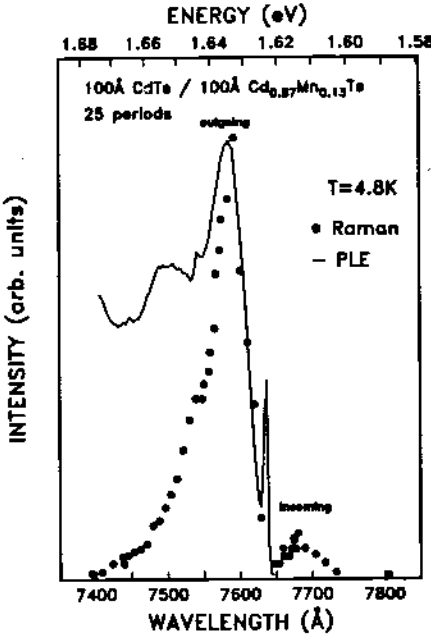

Figure 4: Excitation spectrum of the same sample as in Fig. 1. The points show the Resonance Raman profile of the CdTe-LO phonon, shifted by a phonon frequency.

Finally, we should mention that, since no appreciable shift has been measured in the LO phonon, the CdTe layers are strain-free. The strain in the $Cd_xMn_{1-x}Te$ layers is more difficult to assess from our Raman measurements, since the Mn composition has been estimated only from growth parameters.

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