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RESONANT RAMAN SCATTERING AND EXCITON-PHONON INTERACTION IN CdTe/(Cd,Mn)Te QUANTUM WELLS

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The effect of Raman resonant scattering has yielded much insight into the electronic and vibronic properties of superlattices and quantum wells. Among the former, RRS near excitonic resonances in GaAs/(Ga,Al)As quantum wells has yielded detailed information about the electronic states in this system [1]. We report here on initial resonant Raman studies in CdTe/(Cd,Mn)Te quantum wells with longitudinal optical (LO) phonons near the n=1 ground state exciton [2]. We also note recent similar work by L. Vina and co-workers [3] as reported in this conference. Recent luminescence and excitation spectroscopy [4], which takes advantage of the large g-factors associated with diluted magnetic semiconductors such as (Cd,Mn)Te, has been used to calculate that valence band offsets are small in this system, typically on the order of the exciton binding energy (∼20 meV) [5]. This creates an unusual case for the RRS process in a quantum well where the exciton state is composed from a quasi-2D electron and a quasi-3D hole. As a consequence, the RRS spectra show a number of intriguing and puzzling features as discussed below. At resonance with the n=1 HH exciton the Raman cross section is enhanced over three orders of magnitude correlating well with excitation spectra (the heavy hole notation refers to mass perpendicular to layer plane. |m_z|=3/2 at k=0). However, at the n=1 LH and HH exciton resonances the Raman spectra also show striking broadening and damping effects of the alloy phonon modes. This behavior appears to be a direct consequence of their coupling to the quantum well localized exciton.

The emphasis in these experiments was on a (001) oriented CdTe/(Cd,Mn)Te multiple quantum well (MQW) sample, grown by molecular beam epitaxy (MBE) on (001) GaAs substrate (6). The MQW portion of the structure contained 30 periods of CdTe and (Cd,Mn)Te with Mn alloy fraction x=0.24 in layer thickness ratio approximately 50A/96A. The results reported here were qualitatively very similar to those obtained in another MQW sample with a 84 A CdTe well thickness. For bulk CdTe, the low temperature (excitonic) bandgap is at 1.596 eV and that for the alloy is approximately 382 meV larger. Uniaxial component of lattice mismatch strain is the dominant factor in removing the heavy-light hole valence band degeneracy. The Raman experiments were performed with a low power tunable dye laser (P<5mW) loosely focussed on the samples. The use of a microchannel plate photomultiplier detector increased the versatility of the detection system but limited spectral resolution to about 4 cm⁻¹. The backscattering geometry was z(x',x')L with x' referring to the <110> direction where forbidden LO-processes dominated the Raman cross section.

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The Raman spectrum at incident photon energies above the bandgap of the (Cd,Mn)Te barrier layers was dominated by the CdTe and the MnTe-like modes in the alloy barrier at 167 cm\(^{-1}\) ('LO\(_1\)'), and 198 cm\(^{-1}\) ('LO\(_2\)'), respectively as shown in Fig. 1a. Such two-mode behavior of optical phonons in (Cd,Mn)Te has been elucidated in detail by Ramdas and co-workers both in bulk material (7) and in (Cd,Mn)Te superlattices with emphasis on the study of phonon confinement effects (8),(9). Excitation below the barrier bandgap at 1.757 eV but above the n=1 exciton region added the CdTe LO mode (at 173 cm\(^{-1}\)) to the Raman spectrum indicated by the arrow in Fig. 1b). Figure 1c shows the spectrum obtained at direct resonance within either the n=1 HH or LH exciton absorption line. The spectrum is superposed on a broad luminescence background from direct exciton recombination and shows a striking broadening of the LO phonon lines (to about 25 cm\(^{-1}\)) and a severe damping of the LO\(_2\) mode. The LO\(_1\) mode is broadened so as to partially overlap the CdTe LO-mode. In the resonant 'region' the scattered signal also contained a large depolarized component. At the temperature was raised to 77 K, the alloy modes sharpened considerably under resonant conditions. Raman spectra was also measured for the second order optical phonon spectra where dominant combination tones contained the LO\(_1\) and LO\(_2\) alloy modes. At low temperature and at the n=1 excitonic resonances, large broadening and damping of the two phonon lines was also observed.

The Raman efficiency was strongly enhanced near the n=1 exciton state, as shown in Figure 2 (log scale) for the LO mode. For comparison, the photoluminescence excitation (PLE) spectrum (linear scale) for the same MQW sample is included (10). Additional fine structure apparently related to phonon sidebands are not included in the figure. Both spectra contain the principal n=1 excitonic transitions, namely that of a HH resonance at 1.688 eV and LH resonance at 1.732 eV (separated mainly by the <001> directed uniaxial lattice mismatch strain). For this MQW sample, the n=2 electron confined state is only weakly bound and its interband transition energy is well above the energy range of the figure. For the second MQW sample of 84 A wide CdTe wells, a distinct n=2 HH exciton state was also observed in the RRS spectra, matching well the PLE spectra reported earlier (10). In the RRS spectrum both incoming and outgoing resonances appear near the n=1 'HH' transition although their energy separation surprisingly deviates from the LO-energy by being distinctly smaller. The incoming resonance for the LH state is also present while the outgoing feature has nearly disappeared. These features were reproduced at T= 77 K where, however, the HH incoming-outgoing energy difference is now better matched with the LO energy. Finally, Figure 3 shows the effect of a 4 Tesla external magnetic field on the RRS and excitation spectra in 'Faraday' and 'Voigt' geometries.

The asymmetry in the amplitudes of the outgoing and incoming resonance can be due to scattering by imperfections such as impurities as discussed by Menendez and Cardona (11) or, more likely here, due to the localized nature of the excitons at low temperature. The unusual case of a quasi-2D electron and quasi-3D hole which form the n=1 excitonic states is likely to be responsible for the puzzlingly reduced energy separation between the incoming and outgoing resonances at low temperatures. For the n=1 HH exciton the external magnetic field splits the excitatio spectra as expected in the Faraday geometry [5]; the RRS spectrum shows the corresponding splits at the incoming and outgoing resonances. The case for the LH exciton is considerably more complicated as the excitation and RRS spectra show much less correlation. These details will be considered elsewhere.

Turning back to the phonons, the presence of the barrier alloy phonons in our Raman spectra under direct excitation into the CdTe wells supports the arguments and calculations made for the small valence band offset above. In this interpretation, the hole-LO phonon scattering due to the Frohlich interaction is assumed to be the responsible Raman mechanism. The problem is difficult quantitatively, however, for two main reasons. First, the details of the Raman amplitudes may include partial cancellation effects in a first order Frohlich process in the case of a large difference in energy and hole confinement. Second, the valence bands of CdTe and (Cd,Mn)Te probably include substantial 'HH' and 'LH' coupling for the exciton states.

The striking broadening of the LO\(_1\) alloy mode and the amplitude reduction of the LO\(_2\) mode under resonant excitation (Fig. 1c) was found to be absent above the exciton resonances and at higher temperatures; furthermore, the alloy modes sharpen substantially between the 'HH' and 'LH' resonances within a few meV interval about 1.715 meV. We speculate that the observed effect is connected to the two mode character of the (Cd,Mn)Te alloy in the presence of an additional boundary condition presented by the CdTe heterointerface. Under conditions of extreme resonance, an incident photon couples to excitons which are weakly localized with respect to motion in the layer plane. The exciton-phonon interaction of the alloy modes is thereby limited to a region rather near the heterointerface (probably less than ten lattice constants) and the q=0 wavevector selection rule is relaxed. The exciton may localize weakly as a quasiparticle or, as appears more likely in the CdTe/(Cd,Mn)Te MQW's, be susceptible to hole traps near interfaces. Resonant Raman scattering
Figure 1: Raman spectrum of the LO-phonon at $T=10$ K under excitation (a) above the (Cd,Mn)Te barrier bandgap (showing the two mode alloy behavior; (b) into the CdTe wells but above the $n=1$ exciton resonances (with arrow indicating the LO mode of the CdTe well) and (c) at the exact $n=1$ HH or LH exciton resonance.

Figure 2: Comparison of RRS intensity (closed and open circles for $T=10$ and 77K, respectively) and luminescence excitation spectra at $T=10$ K (solid line). The energy scales at top and bottom are for 10 K and 77 K, respectively. The dashed lines through RRS spectra are to guide the eye. The RRS intensity is on log scale while that for PLE is linear.

from 3D excitons localized at impurities has been studied in doped compound or mixed crystal semiconductors with clear indications of the presence of additional local vibrational modes [12]. Here, particularly for the MnTe-like LO2 mode, the CdTe interface presents a boundary condition which disallows its excitation at the geometrical interface i.e. the large reduction in the effective Mn-ion concentration within about a lattice constant from the interface changes the character of the MnTe vibrations to an increasingly 'local' one.

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References


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Figure 3: Comparison of RRS and PLE intensities in the presence of a 4 Tesla magnetic field parallel to the superlattice axis (left) and perpendicular to z-axis (right). The arrows in the left panel indicate the spin splitting of the incoming and outgoing resonances of the HH ($|m_j|=3/2$) exciton.