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Raman and resonant Raman scattering from the HgTe/CdTe superlattice

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(Received 11 August 1989)

Raman spectra from two high-quality HgTe/CdTe superlattices (SL's) [HgTe/CdTe, (80 Å)/(40 Å) and (64 Å)/(60 Å)] show HgTe, CdTe, and $Hg_{1-x}Cd_xTe$ phonon modes. Our results and earlier Raman data [Olego, Faurie, and Raccah, Phys. Rev. Lett. **55**, 328 (1985)] show that outgoing resonance with the CdTe $E_0+\Delta_0$ gap is important, complicating the study of SL effects. An unknown broad peak near 430 cm^{-1} is similar to a feature reported by Olego, Faurie, and Raccah but is not due to photoluminescence. Another unknown Raman band is seen at 155 cm^{-1}. The locations of the $Hg_{1-x}Cd_xTe$ phonon lines confirm the degree of Hg alloying in the nominal CdTe layers, showing the utility of Raman scattering for SL characterization.

I. INTRODUCTION

The proposal of the HgTe/CdTe superlattice (SL) as a new material for infrared optoelectronics, with advantages compared to the $Hg_{1-x}Cd_xTe$ alloy of the same band gap, has led to much effort in making and understanding this II-VI compound microstructure.[1-4] Despite this intense interest, however, only one Raman scattering study has appeared, that of Olego, Faurie, and Raccah (OFR).[5] Here we present new and thorough Raman and resonant Raman scattering results from two high-quality HgTe/CdTe SL's grown by molecular-beam epitaxy (MBE).

The earlier Raman results, with photoluminescence (PL) data, were interpreted as showing shifts in the $E_0+\Delta_0$ (Γ_6-Γ_7) edge due to SL confinement. A striking feature of these spectra was that although multiple harmonics of the CdTe longitudinal-optical (LO) phonons appeared, no features connected with HgTe were seen. In contrast, we see phonon modes from both CdTe and HgTe, as well as CdTe-like and HgTe-like modes from alloy $Hg_{1-x}Cd_xTe$. Our recent thorough study of outgoing resonance and PL in epitaxial CdTe films[6] explains many of our observations and offers an alternate explanation for the earlier Raman data.

II. EXPERIMENT

The HgTe/CdTe SL's were grown in a commercial MBE system at Hughes Research Laboratories. Alternating CdTe and HgTe layers were deposited at rates of 4 and 3.2 Å/sec, respectively, on 3.6-μm CdTe buffer layers grown on high-quality (x-ray double crystal rocking curves half-widths $<20''$) commercial (100) CdTe substrates. Growth temperature was 170 °C and source pressures were typically 1.7×10^{-4}, 1.4×10^{-6}, and 1.6×10^{-6} mbar for Hg, Te_2, and CdTe sources, respectively. Superlattice no. 2 has a 300-Å CdTe cap for surface passivation atop 150 periods of [HgTe (80 Å)]/[CdTe (40 Å)], whereas SL no. 1 has 150 periods of [HgTe (64 Å)]/[CdTe (60 Å)] with no CdTe cap.

The Hg beam was left on throughout growth because the pumping time for Hg is long compared to that for Te_2 and CdTe. Thus, Hg is incident on the SL's even during CdTe growth and the nominal CdTe layers become $Hg_{1-x}Cd_xTe$ with $x\simeq0.85$, as shown by x-ray diffraction, energy-dispersive x-ray analysis, and infrared (ir) photoluminescence (PL) (Ref. 7).

The SL's were of high quality, as indicated by cross-sectional transmission electron microscopy (XTEM), which showed abrupt interfaces between the HgTe and $Hg_{1-x}Cd_xTe$ layers, Hall data, which gave high carrier mobilities, and room-temperature transmission and reflection measurements, which displayed heavy- and light-hole interband transitions.[8] Far-infrared spectroscopy[9] at Emory confirmed these findings.

Raman spectra were obtained at Emory in the near-backscattering geometry at sample temperatures of 10 and 80 K. The samples were mounted on a cold finger and excited by an Ar^+ laser with focusing spot size <0.3

mm. Resonant Raman effects were studied with eight laser lines covering 2.4–2.7 eV. The scattered light was dispersed by a triple spectrometer and detected by an optical multichannel analyzer (OMA), which gave high sensitivity with a resolution of 2–3 cm^{-1}.

III. ANALYSIS

Figure 1 shows Raman spectra for SL no. 1, without a cap layer, and SL no. 2, with a CdTe cap layer, at 10 K and at 476.5 nm, with line assignments given in Table I. Unlike the spectra obtained by OFR at the same wavelength and nearly the same temperature, which showed only CdTe lines with no evidence of HgTe, our result for SL no. 1 [Fig. 1(a)] shows HgTe-related lines with virtually no CdTe lines.

The HgTe transverse-optical (TO) and longitudinal-optical (LO) phonon peaks are prominent at 118 and 138 cm^{-1}, and the HgTe-like LO line from $Hg_{0.15}Cd_{0.85}Te$ appears at 132 cm^{-1} (peak 2). The HgTe-like TO mode at 127 cm^{-1} is submerged in the low-energy tail of the pure HgTe and HgTe-like LO bands. Second-order phonon combinations appear over 240–340 cm^{-1}, where the strongest feature at 278 cm^{-1} is the HgTe 2LO mode (peak 6). A broad feature (labeled *G*) extends from 350 to 550 cm^{-1}, with a peak near 430 cm^{-1}; OFR reported a similar feature. The only evidence of CdTe phonons is the combined CdTe LO+HgTe-like LO line at 295 cm^{-1} (peak 7).

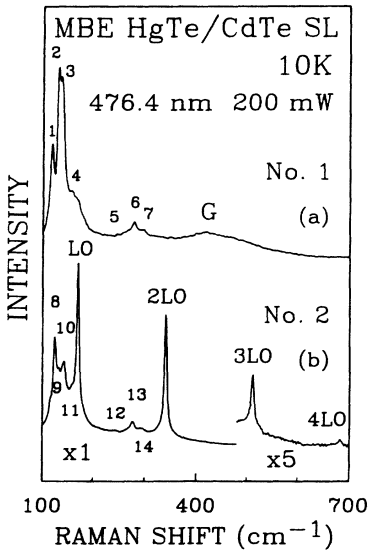

FIG. 1. Raman spectra of two HgTe/CdTe superlattices at 10 K, excited at 476.4 nm. (a) SL sample no. 1, with 150 periods of [HgTe (64 Å)]/[CdTe (60 Å)] and with no CdTe cap. (b) SL sample no. 2, with 300-Å CdTe cap, 150 periods of [HgTe (80 Å)]/[CdTe (40 Å)], and 3.6-μm CdTe buffer, on CdTe substrate. Labeled line positions and assignments are given in Table I. Line intensities in (a) are not comparable to those in (b) because each plot uses its own arbitrary units.

The apparent contradiction between the dominant HgTe phonons in our data and the dominant CdTe phonons in OFR's data can be resolved in terms of resonant effects. If the exciting Raman light resonates with an energy gap characteristic of one SL layer type but not the other, the corresponding phonon peaks can be enhanced sufficiently to overwhelm the peaks from the other layer type. The weaker phonon set is especially difficult to see with a high luminescent background or low signal-to-noise ratio. Our data, obtained with a triple spectrometer and OMA detection, give superior resolution of small structure.

A careful study of OFR's spectra suggests that outgoing resonance with the $E_0+\Delta_0$ gap in CdTe is a likely cause for the appearance of CdTe peaks only. In OFR's Raman spectra from three different SL's at 476.5 nm (see OFR, Fig. 1), the 2LO and 3LO lines are much larger than the 1LO and 4LO peaks. In two cases the 3LO line is larger than the 2LO line. This is very similar to what we see in our extensive study of outgoing resonance Raman scattering near the $E_0+\Delta_0$ gap in CdTe films.[6]

Outgoing resonance occurs when the energy E_{in} of the incoming photon, after it has been scattered by n phonons, lies near an energy gap E_g (Refs. 6 and 10). For LO phonons of frequency ω_{LO}, the resonant condition is

$$E_{in}-n\hbar\omega_{LO}=E_g \ . \tag{1}$$

Our study of CdTe showed that the nth LO harmonic and its neighboring $(n\pm1)$th harmonics are strongly enhanced relative to the other peaks. We use the known value $\hbar\omega_{LO}=21.6$ meV in CdTe and assume that E_g is the $E_0+\Delta_0$ gap in CdTe to obtain n from Eq. (1). OFR give $E_0+\Delta_0=2.530$ eV for a CdTe film, and state that confinement effects shift this value up to 2.540 eV for the CdTe layers in their SL. These two values give $n=3.3$ and 2.8, respectively, both reasonably close to an integer and consistent with the observation that the 2nd and 3rd overtones are enhanced in OFR's data. Our calculation does not distinguish confined states from nonconfined ones.

To show how increased strength in the CdTe modes minimizes the HgTe peaks, Fig. 1(b) gives the Raman spectrum at 476.5 nm of SL no. 2, with its CdTe cap layer. Alloy HgTe-like modes like those in no. 1 still appear for SL no. 2. Peak 8 (126 cm^{-1}) and peak 9 (133 cm^{-1}) lie at the HgTe-like TO- and LO-phonon frequencies, respectively, from $Hg_{0.15}Cd_{0.85}Te$. (To further confirm the presence of the alloy, the CdTe-like LO mode from $Hg_{0.15}Cd_{0.85}Te$ appears as expected at 165 cm^{-1} (peak 11), as a shoulder of the strong pure CdTe LO band.) The CdTe-like TO mode, which should appear near 144 cm^{-1}, is submerged by the pure CdTe TO band (peak 10, 143 cm^{-1}). However, the HgTe-like modes are dwarfed by the CdTe LO and 2LO modes. The latter may be large simply because the CdTe cap layer absorbs much of the incoming light before it reaches the SL; however, the relative strength of the CdTe 2LO mode, as well as spectra of sample no. 2 measured at other wavelengths, suggest that outgoing resonance with $E_0+\Delta_0$ occurs in the

TABLE I. Raman assignments for HgTe/CdTe superlattices no. 1 and no. 2. See Fig. 1.

SL sample no.	Line	Raman shift (cm^{-1})	Assignment
1	1	118	HgTe TO
	2	132	HgTe-like LO (with HgTe-like TO)
	3	138	HgTe LO
	4	155	?
	5	265	HgTe-like 2LO
	6	278	HgTe 2LO
	7	295	CdTe LO+HgTe-like LO
	G	425	?
2	8	126	HgTe-like TO
	9	133	HgTe-like LO
	10	143	CdTe TO (with CdTe-like TO)
	11	165	CdTe-like LO
	LO	171	CdTe LO
	12	244	?
	13	277	HgTe-like LO+CdTe TO or HgTe-like LO+CdTe-like TO
	14	296	CdTe LO+HgTe-like TO
	2LO	341	CdTe 2LO
	3LO	512	CdTe 3LO
	4LO	683	CdTe 4LO

CdTe cap as it did in our CdTe films[6] to further amplify the CdTe modes.

Returning to sample no. 1 with its broad peak *G* at 430 cm^{-1}, OFR interpreted a similar peak in their spectra as due to photoluminescence across the $E_0+\Delta_0$ gap.[5] Later Olego and Faurie (OF) further studied the E_0 and $E_0+\Delta_0$ edges in HgTe/CdTe heterostructures by photoluminescence spectroscopy.[11] OFR and OF never observed any similar broad peak from a CdTe film alone. OFR concluded that the peak came from SL effects related to the quantization of the valence Γ_7 band in the CdTe layers, and from this determined that confinement effects shifted up the $E_0+\Delta_0$ edge by several meV.[5,11]

However, our study of CdTe films has shown clear PL across the $E_0+\Delta_0$ gap at temperatures up to 80 K, sometimes together with Raman peaks, provided there were sufficient carriers.[5] Hence a peak at or near $E_0+\Delta_0$ is not a unique SL effect. Further, Figs. 1 and 2 show that peak *G* is not due to PL at all because its Raman shift is independent of excitation wavelength over 457.9–514.5 nm (although its intensity relative to first-order features does change with wavelength). The peak may well be connected to SL properties such as carrier confinement in the HgTe wells, because it appears for SL no. 1, with a HgTe layer thickness of 6.4 nm, but not for SL no. 2, with a wider HgTe layer of 8.0 nm. Sample SL no. 1 also exhibits an unknown feature at 155 cm^{-1} [Figs. 1(a) and 2], far from any known CdTe, HgTe, or $Hg_{0.15}Cd_{0.85}Te$ modes, which may also come from the SL structure.

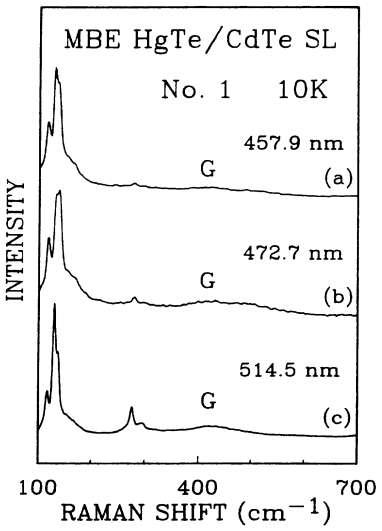

FIG. 2. Raman spectra of HgTe/CdTe superlattices no. 1 at 10 K, at three excitation wavelengths.

IV. CONCLUSIONS

We have measured and analyzed Raman spectra at several wavelengths from two well-characterized, high-quality HgTe/CdTe SL's. Our prime observation, in the SL without a CdTe cap, is that our data seem to contradict the results of OFR. We see virtually no evidence of CdTe modes, whereas OFR report no HgTe modes. Our detailed study of outgoing resonance in CdTe, and our analysis of OFR's data, however, show that the HgTe modes are not mysteriously lost in OFR's spectra; they

are only too small to appear relative to CdTe modes made very intense by outgoing resonance with the $E_0+\Delta_0$ gap in the CdTe layers.

This alone does not explain why outgoing CdTe resonance does not also occur in our sample SL no. 1 to dominate the HgTe modes. One possibility is that in SL no. 1, there is a resonance with a HgTe gap energy which causes the HgTe and HgTe-like phonons to dominate the CdTe phonons. In fact, Raman spectra for SL no. 1 at different wavelengths (not shown here) suggest an incoming resonance with the HgTe E_1 gap near 2.3 eV at low temperature.

Our discussion focuses on higher-order gaps associated with the individual SL layer materials, possibly shifted by confinement. We recognize that similar effects could well arise from gaps associated with full SL energy states. However, detailed calculations of resonant Raman scattering are difficult even in bulk material. We have not attempted to develop the difficult equivalent theory for the detailed higher-order HgTe/CdTe SL energy states. Nevertheless, our first-order approach does show that any interpretation of SL Raman spectra must include complicating resonance effects. Very careful resonance Raman experiments, using continually tunable radiation from a dye laser, could accurately probe gap energies and hence perhaps distinguish between confined and nonconfined states or between layer and SL states.

One intriguing feature, the broad peak at 430 cm^{-1}, is definitely not due to PL, in contradiction to the reports of OFR and OF, and may represent a SL effect. The 155-cm^{-1} mode we see in SL no. 1 also has no obvious explanation.

Our Raman analysis gave ample evidence that the CdTe layers also contained Hg, as expected by the grower and confirmed by other methods. The resolution of our measured Raman frequencies for HgTe-like and CdTe-like modes from the $Hg_{1-x}Cd_xTe$ layers is sufficient to establish the x value as 0.84 ± 0.03, confirming the value ~ 0.85 obtained by other measurements. Hence Raman scattering can be useful to characterize layer alloying in HgTe/CdTe and, presumably, other superlattices.

ACKNOWLEDGMENTS

We thank F. A. Shirland for technical help and L. S. Kim for useful discussions.

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