Composition dependence of longitudinal optical phonon modes in $Cd_xHg_{1-x}Te$ with $0.5 \leq x \leq 1$

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Using Raman scattering, we have studied the composition dependence of the CdTe-like longitudinal optical (LO) phonon mode in $Cd_xHg_{1-x}Te$ with $0.5 \leq x \leq 1$. For excitation with photon energies close to the $E_0+\Delta_0$ gap energy, a resonance enhancement was found at 77 K for dipole-forbidden Fröhlich-induced one-LO-phonon and Fröhlich-induced two-LO-phonon scattering over the whole composition range. Taking advantage of this enhancement we determined the one-LO-phonon and two-LO-phonon frequencies and found a frequency shift linear with x. This shift is consistent with a linear composition dependence of the CdTe-like LO mode over the whole range from CdTe to HgTe.

$Cd_xHg_{1-x}Te$ mixed crystals can be formed over the whole composition range from $x=0$ (HgTe) to $x=1$ (CdTe). The composition dependence of the longitudinal optical (LO) and transverse optical (TO) phonon frequencies in these alloys has been studied so far mainly using infrared reflectivity measurements.[1-4] A two-mode behavior was found with CdTe-like and HgTe-like LO- and TO-phonon modes, respectively. Only in two studies Raman spectroscopy was used to analyze the composition dependence of the lattice dynamics of $Cd_xHg_{1-x}Te$.[4,5] Recently Menéndez *et al.*[6] investigated the resonance behavior of dipole-forbidden Fröhlich-induced one-LO-phonon and Fröhlich-induced two-LO-phonon Raman scattering for incident photon energies close to the $E_0+\Delta_0$ band-gap energy. They found at 77 K a sharp $E_0+\Delta_0$ resonance in pure CdTe which gets broadened and reduced in peak height with increasing Hg content ($0.944 \leq x \leq 1.0$). All the work mentioned so far was carried out on bulk grown $Cd_xHg_{1-x}Te$ samples.

The purpose of the present study was to investigate the composition dependence of the CdTe-like one- and two-LO-phonon modes in high-quality Cd-rich $Cd_xHg_{1-x}Te$ epitaxial layers. Raman spectroscopy was used with the excitation close to resonance with the $E_0+\Delta_0$ band-gap energy to take advantage of the resonance enhancement of the Raman scattering efficiency.

The samples used for this study were grown by liquid-phase epitaxy (LPE) onto (111) $Cd_{0.96}Zn_{0.04}Te$ substrates.[7] The layers are about 10 μm thick and they have a very good compositional homogeneity. The lateral and in-depth fluctuations of composition are less than 0.5 mol % CdTe.

The Raman spectra were excited with several lines of a Kr-ion and an Ar-ion laser. The samples were cooled to 77 K and the spectra were recorded in backscattering from a (111) surface with the scattered light not analyzed for its polarization. For these conditions, dipole-allowed and dipole-forbidden but Fröhlich-induced one-LO-phonon scattering are observed. But at low temperatures and for excitation close to the $E_0+\Delta_0$ gap energy the latter scattering mechanism is dominant.[6] The incident power on the samples was <50 mW. The scattered light was filtered and dispersed in a triple monochromator and detected with an intensified silicon diode array. The spectral resolution varied between 3.5 and 5 cm^{-1}.

Figure 1 displays a sequence of Raman spectra of a $Cd_xHg_{1-x}Te$ sample with $x=0.71$ excited at different photon energies. For excitation at 2.38 eV the CdTe-like LO-phonon mode is observed at 164.8 cm^{-1} together with the HgTe-like LO-phonon mode at 133 cm^{-1}. In between lies the CdTe-like TO-phonon mode at ~ 143 cm^{-1} which is hardly resolved in this spectrum. The two latter modes overlap with two additional Raman lines at ~ 128 and 143 cm^{-1} which have been ascribed to excess Te on the sample surface.[8] Excitation at 2.18 and 1.91 eV results in a drastic enhancement of the CdTe-like LO-

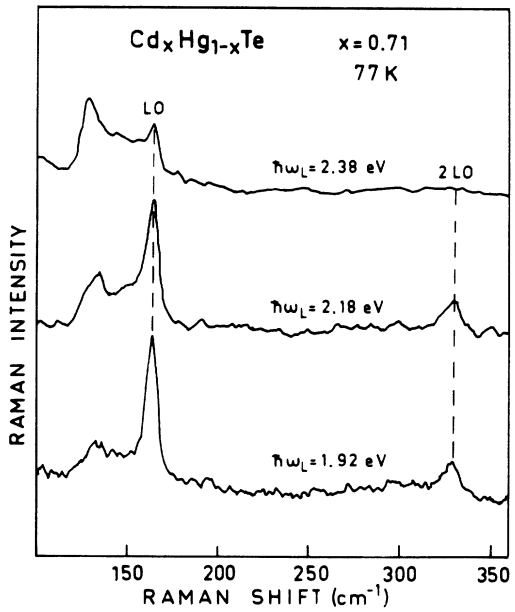

FIG. 1. Low-temperature Raman spectra of $Cd_{0.71}Hg_{0.29}Te$ excited at three different photon energies indicated in the figure.

phonon line with respect to the CdTe-like TO- and HgTe-like LO-phonon modes. In addition, scattering by two CdTe-like LO phonons is seen at 329.5 cm^{-1}.

According to Menéndez *et al.*,[6] the resonances of scattering by one and two CdTe-like LO phonons lie at energies which are shifted to higher energies with respect to the $E_0+\Delta_0$ band-gap energy by one and two LO-phonon energies, respectively (outgoing resonance). Using the semiempirical formula given by Legros and Triboulet[9] to calculate the band-gap energy E_0 for a given composition and temperature and taking a composition- and temperature-independent value of $\Delta_0=0.921$ eV (Ref. 10) we calculated the one- and two-LO-phonon resonance energies to be 1.913 and 1.934 eV, respectively, for $x=0.71$ at 77 K. This is in agreement with the experimental data shown in Fig. 1 where the strongest resonance enhancement of the CdTe-like LO-phonon lines, relative to the CdTe-like TO- and HgTe-like LO-phonon modes, is found for excitation at 1.92 eV. It is interesting to note that no such enhancement occurs for the HgTe-like LO-phonon mode.

Figure 2 shows a series of Raman spectra which were recorded from samples with different compositions but excited at a fixed photon energy of 2.38 eV. A resonance enhancement of the CdTe-like one- and two-LO-phonon modes is found with a maximum for $x=0.92$. The corresponding resonance energies for scattering by one and two LO phonons are 2.342 and 2.363 eV, respectively, which is again close to the incident photon energy of 2.38 eV.

The above results show that even for fairly large Hg contents (e.g., $x\simeq 0.7$), where alloying effects are expected to broaden that $E_0+\Delta_0$ resonance, a pronounced resonance enhancement of the dipole-forbidden one- and two-LO-phonon Raman scattering is found. This enhancement allows a clear separation of the CdTe-like LO-phonon mode from the other vibrational modes over the whole composition range ($0.5\leq x\leq 1.0$) studied here. For that reason, and also because the CdTe-like TO-phonon mode and the HgTe-like LO-phonon mode are interfering with Te modes,[8] we concentrate in the following on the composition dependence of the CdTe-like one- and two-LO-phonon modes.

Figure 3 displays the Raman shift of the one- and two-LO-phonon lines plotted versus the composition x. The data were extracted from Raman spectra excited at different photon energies indicated in the figure. The straight lines show the variation of the one- and two-LO-phonon frequencies with x expected for a linear change of the mode frequencies between the values for CdTe and HgTe as reported by Baars and Sorger.[1] This work was based on infrared reflectivity measurements on Hg-rich $Cd_xHg_{1-x}Te$ samples. Also, recent reflectivity measurements by Hoclet *et al.*[2] on Cd-rich alloys are consistent with such a linear shift. Our present Raman data are, within the experimental scatter, in good agreement with the above cited reflectivity data.[1]

Combining Raman scattering and infrared reflectivity, Vodopyanov *et al.*[4] reported a composition dependence of the CdTe-like LO-phonon frequency which shows a smaller change with x for $x>0.4$ as one would expect from a linear interpolation between the corresponding

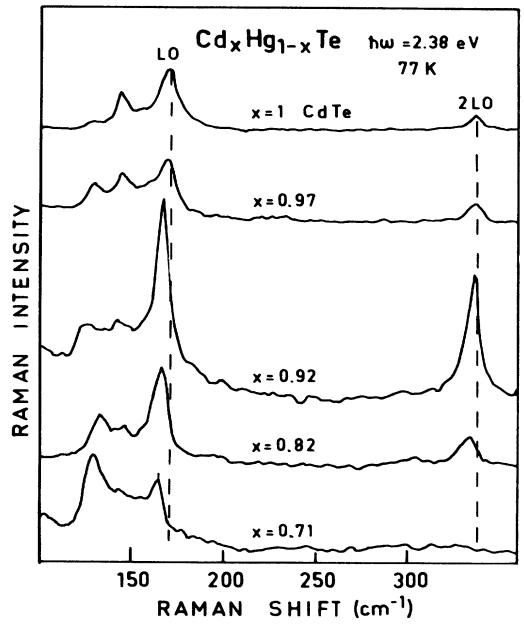

FIG. 2. Low-temperature Raman spectra of $Cd_xHg_{1-x}Te$ for various compositions x indicated in the figure. The spectra were excited at 2.38 eV. The dashed lines indicate the one- and two-LO-phonon frequencies in CdTe.

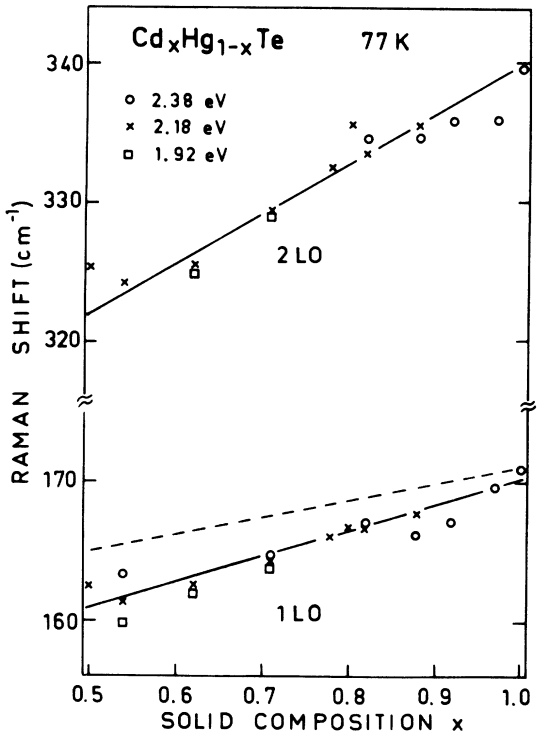

FIG. 3. Frequency of the one- and two-LO-phonon Raman lines vs solid composition x measured at 77 K. Different incident photon energies are indicated in the figure. The drawn curves are 77 K data from Ref. 1 with the values for the two-LO-phonon curve obtained by multiplying by 2. The dashed curve represents the data from Refs. 3 and 4.

frequencies in CdTe and HgTe. Their results are shown in Fig. 3 by the dashed line. They attribute this peculiar behavior to strong electron-phonon interactions for alloy compositions, for which the E_0 band-gap energy comes close to the CdTe-like LO-phonon energy.[3] As it is seen from Fig. 3, the present data do not support the results by Vodopyanov *et al.*[4] but favor the infrared reflectivity data of Baars and Sorger[1] and of Hoclet *et al.*[2]

To summarize, Raman scattering with excitation resonant with the $E_0+\Delta_0$ band gap has been used to study the composition dependence of the CdTe-like LO-phonon modes in Cd-rich $(0.5 \leq x \leq 1.0)$ $Cd_xHg_{1-x}Te$. For these conditions a resonance enhancement of the dipole-forbidden one- and two-LO-phonon scattering was observed at 77 K over the whole composition range investigated. A linear shift of the one- and two-LO-phonon frequencies was found as described by a linear variation of the mode frequencies between the corresponding frequencies in the two binary compounds CdTe and HgTe.

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