Raman scattering by optical phonons in $In_{1-y-z}Al_yGa_zAs$ lattice matched to InP

R. Borroff and R. Merlin
Department of Physics, The University of Michigan, Ann Arbor, Michigan 48109-1120

A. Chin and P. K. Bhattacharya
Department of Electrical Engineering and Computer Science, The University of Michigan, Ann Arbor, Michigan 48109-2122

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We report on Raman scattering by longitudinal optical phonons in $In_{1-y-z}Al_yGa_zAs$ ($1-y-z=0.53$) lattice matched to InP. The quaternary alloys were grown on (001) InP by molecular beam epitaxy. The phonon spectra exhibit three-mode behavior. The frequencies of AlAs- and GaAs-like modes vary linearly with the concentration of Al (or Ga) while the position of the InAs-like phonon remains nearly constant. The data show no evidence of alloy clustering.

The $In_{1-y-z}Al_yGa_zAs/InP$ lattice-matched system holds promise for device applications relevant to optical communications. This is mainly because the band-gap range covered by the lattice-matched quaternary alloys overlaps the ≈ 1.1–$1.5\,\mu$m (0.8–1.1 eV) region of minimum loss and dispersion for current optical fibers [at 300 K, the gap varies from 0.74 eV ($In_{0.53}Ga_{0.47}As$) to 1.45 eV ($In_{0.52}Al_{0.48}As$)]. Growth of $In_{1-y-z}Al_yGa_zAs/InP$ ($1-y-z\approx 0.52$–0.53) by molecular beam epitaxy (MBE) has been reported recently.[1–6] The use of MBE, as opposed to, e.g., liquid phase epitaxy (LPE), is favored by the fact that all group III elements show nearly the same ($\cong 1$) sticking coefficient. Parenthetically, we note that the growth of these alloys by LPE is rendered difficult because of the large distribution coefficient of Al in the liquid phase.[7] Compared to $In_yGa_{1-y}As_zP_{1-z}$ lattice matched to InP, which also covers the energy range of interest for optical communication, MBE growth of $In_{1-y-z}Al_yGa_zAs$ presents the advantage of avoiding the need for a stringent control of group V element ratio (as well as eliminating the use of P). Problems such as clustering[7] originated in the presence of miscibility gaps in the solid phase (as for the end ternary $In_{0.52}Al_{0.48}As$) can be overcome to some extent by choosing the appropriate set of growth parameters. However, unequal (In-As, Ga-As, and Al-As) bond strengths and associated adatom mobilities controlling MBE growth can easily lead to substantial interface roughness. This problem is also shared by the growth of ternary alloys, but it is clearly more difficult to solve in the quaternary case.

In this letter, we report on a Raman scattering (RS) study of the alloy composition dependence of longitudinal optical (LO) phonons in $In_{0.53}(Al_xGa_{1-x})_{0.47}As$ lattice matched to InP. An important outcome of our investigation is that RS provides a simple and accurate way to characterize the alloys. Extensive Raman studies have shown this to apply to other III-V quaternaries[8] and ternaries[9–11] as well. In particular, the analysis of our results relies largely on previous work on the alloys $In_yAl_{1-y}As$[9] and $In_yGa_{1-y}As$.[10,11] The latter ternaries exhibit two-mode behavior, i.e., their spectra show two LO (as well as two transverse optical) modes associated with the respective end compounds for nearly the whole composition range.[9–11] Our Raman data on $In_{0.53}(Al_xGa_{1-x})_{0.47}As$ reveal three-mode behavior. The x dependence of the AlAs- and GaAs-like LO frequencies is approximately linear. The InAs-like mode does not depend much on x and it may exhibit a weak nonmonotonic component.

The $In_{0.53}(Al_xGa_{1-x})_{0.47}As$ samples investigated in this work were grown by MBE on (001) InP. Layer thicknesses were ≈ 1.5 μm and $x=0$, 0.11, 0.36, 0.64, 1 (± 0.01). The latter values were inferred from growth parameters and results of electron microprobe analysis (x-ray fluorescence). Photoluminescence spectra of the samples reveal (possibly bound) excitonic features with full widths at half-maxima in the range 6.5 meV ($x=0.36$)–21.9 meV ($x=0.64$) at $T=15$ K. Measurements of x-ray scattering give $\Delta a/a\approx 2.5\times 10^{-3}$ ($x=0.64$), $\Delta a/a\approx 3.5\times 10^{-4}$ ($x=0.36$), and $\Delta a/a\leqslant 10^{-4}$ ($x=0.11$) for the lattice mismatch; Δa is the difference between the lattice parameter of the quarternaries (along the growth direction) and a, the corresponding parameter for InP. A detailed account of the growth process and the characterization of the layers will be given elsewhere.

The Raman experiments were performed at $T=300$ K using several lines of Ar^+ and Kr^+ lasers. Spectra were recorded in the $z(x,\ x+y)\bar{z}$ backscattering configuration where z is normal to the layers and x, y are along the [100] and [010] directions. This geometry only allows scattering by LO modes with wave vectors parallel to [001]. The notation $(x,\ x+y)$ indicates that the polarization vector of the incident beam is along [100] and that the scattered photons are not analyzed. Accordingly, the spectra include both the allowed and the so-called forbidden contribution to LO scattering. The best Raman results were obtained with the Ar^+ line at ≈ 2.602 eV (4765 Å) giving the largest measured value of the ratio between LO and disorder-induced phonon intensities (see below). For small x's, this line further leads to enhanced (resonant) scattering due to its proximity to the E_1 gap of $In_{0.53}Ga_{0.47}As$.[11]

Our results are summarized in Figs. 1 and 2. Raman spectra of the five samples are shown in Fig. 1. The data for $x=0$ are in very good agreement with those obtained under

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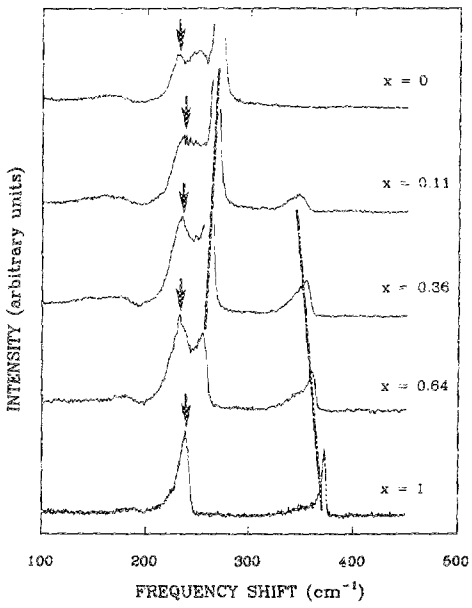

FIG. 1. Raman spectra of $In_{0.53}(Al_xGa_{1-x})_{0.47}As$ at $T = 300$ K. Arrows indicate the position of the InAs-like LO phonon. Dashed lines track with the GaAs-like (at lower frequency shifts) and the AlAs-like LO modes. Other features are disorder-activated phonon bands (see text). The laser energy is 2.602 eV (4765 Å) and the scattering geometry is $z(x, x+y)\bar{z}$.

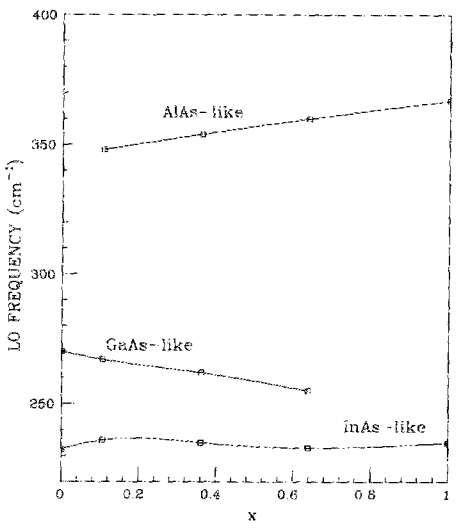

FIG. 2. Frequencies of the LO modes as a function of x (the lines are guides to the eye). The data correspond to the spectra shown in Fig. 1. Estimated errors are ± 0.01 for x and ± 2 cm^{-1} for the LO frequencies.

the same conditions for LPE-grown $In_{0.53}Ga_{0.47}As$.[11] The dominant feature at 270 cm^{-1} corresponds to the GaAs-like LO phonon while the InAs-like LO mode is the weaker peak at 232 cm^{-1}.[10,11] The relatively broader structures at ≈ 150 and 250 cm^{-1} are due to (alloy) disorder-induced scattering[10,11] by, respectively, longitudinal acoustic and optical phonons (we note that the structure at 250 cm^{-1} has been previously ascribed to the GaAs-like transverse mode,[11] but this is inconsistent with our scattering geometry). With increasing x, the disorder-activated features weaken considerably. The acoustic band shows a positive shift; its maximum is at ≈ 182 cm^{-1} for $x = 1$. The spectrum of $In_{0.53}Al_{0.47}As$ ($x = 1$) is very similar to those reported for MBE-grown $In_yAl_{1-y}As$ alloys of nearly the same composition.[9] The Raman peaks at 238 and 272 cm^{-1} correspond to the InAs- and AlAs-like LO modes, respectively.[9] The asymmetry of these peaks, and other LO features in Fig. 1, originates in the scattering by phonons with wave vectors $\mathbf{k} \neq 0$ activated by the alloy disorder.[12] Their line shapes reflect the negative curvature of the LO branch.[12]

The spectra of the quaternaries ($x = 0.11$, 0.36, and 0.64) show that the intensity of the GaAs-like mode (relative to that of the InAs-like phonon) decreases with increasing x. Moreover, there is a new feature appearing at ≈ 350 cm^{-1} ($x = 0.11$) which evolves into the $x = 1$ AlAs-like phonon. These results indicate that the character of the (long-wavelength) phonon spectrum of the quaternaries is of the three-mode type. The measured frequencies of the three LO modes as a function of x are shown in Fig. 2. The InAs-like phonon depends only weakly on x. This is consistent with its weak y dependence in $In_yAl_{1-y}As$ and $In_yGa_{1-y}As$ alloys.[9–11] The behavior in $In_{0.53}(Al_xGa_{1-x})_{0.47}As$ appears to be nonmonotonic, but the frequency changes are too close to the experimental errors (± 2 cm^{-1}) to be certain. The GaAs-like and the AlAs-like LO modes show a nearly linear dependence with composition providing an effective marker for characterization purposes.

The dependence of the spectra on x measures, in some sense, the uniformity of the alloys. The Raman data show no evidence of features attributable to regions of different composition in the layers. This and the results in Fig. 2 indicate that, in the length scale probed by RS ($1/q \approx 1300$ Å; q is the scattering wave vector), clustering is not important in our samples.

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