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Raman spectra from $Ga_{1-x}In_xAs$ epitaxial layers grown on GaAs and InP substrates

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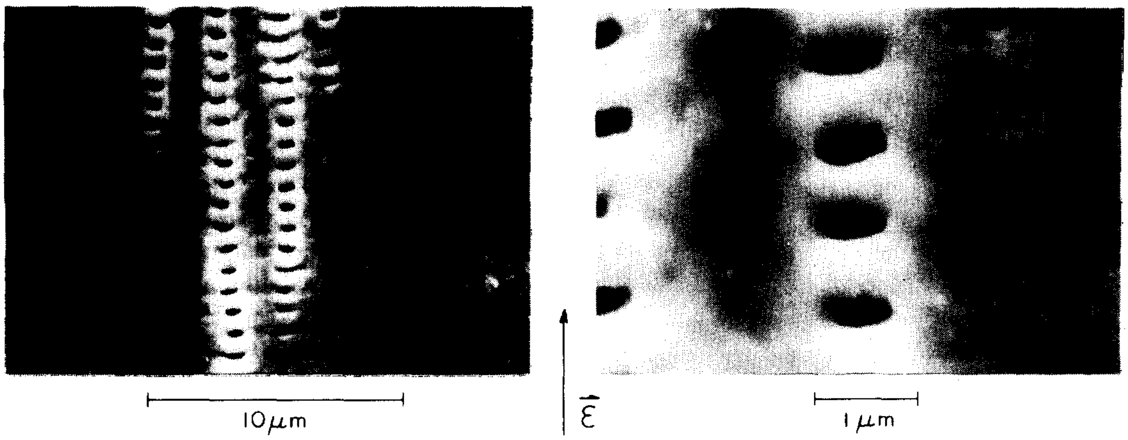

FIG. 5. SEM photographs of ripples produced in GaAs using a single 1.06-μm pulse. The structures shown occur near the center of the ~80-μm-diam laser spot. The ripples run horizontally in the photographs (*E*-field polarization is vertical) and occur in narrow strips, each strip being an array of pits spaced by the ripple spacing.

low the threshold of transition from amorphous to polycrystal, can also create ripples, even when single-shot illumination produces no observable effect.[8]

One complication is that at lower intensities the ripples sometimes tend to be organized in stripes of varying width and spacing. Figure 4 illustrates several of the points just made, for the case of $\lambda = 1.06$-μm illumination of GaAs. The spot size in these experiments was 80 μm in diameter, with the ripples occurring at the center of the spot, where the energy fluence was ≈ 250 mJ/cm^2. These low-intensity ripples in GaAs also exhibit a rather amazing small-scale structure, as shown in the secondary electron microscopy (SEM) closeups in Fig. 5. The origin of these pits is not understood, but they may correspond to small regions at the peaks of the ripples where arsenic has locally evaporated and redeposited around the rim. As secondary evidence we find in GaAs that when the incident intensity is increased above that used in Figs. 4 and 5, no improvement in the electrical properties is observed, while in our silicon samples wherever the intensity is above the melting threshold the sheet resistivity drops below 100 $\Omega/\square$.

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Raman spectra from $Ga_{1-x}In_xAs$ epitaxial layers grown on GaAs and InP substrates

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Raman spectra from $Ga_{1-x}In_xAs$ epitaxial layers of various compositions were studied. Both disorder-activated acoustic and optical phonons appeared in the midrange of composition independent of substrate materials. Broadening in the LO phonon due to stress was also observed near the interface region between the epitaxial layer and the substrate when there was lattice mismatch between them even if the amount was as small as 0.7%.

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$Ga_{1-x}In_xAs$ has recently been of great interest as a useful material for optoelectronic and high-speed devices. Fundamental properties of $Ga_{1-x}In_xAs$ such as disorder in atomic arrangement and stress in the materials, however, have hardly been made clear. For example, Raman spectra from $Ga_{1-x}In_xAs$ have not been discussed sufficiently although those from $Ga_{1-x}Al_xAs$,[1] $InP_{1-x}As_x$,[2] and $Ga_{1-x}In_xP$[3] have been reported in many papers. Raman and infrared absorption spectra are well known to be affected by disorder in ternary alloys. Yamazaki *et al.*[4] reported that long wavelength optical phonons in $Ga_{1-x}In_xAs$ can be explained with a cluster model. Localized modes have been observed in Raman spectra from some ternary alloys such as $Ga_{1-x}Al_xAs$,[5] $InP_{1-x}As_x$,[2] and $In_{1-x}Ga_xP$.[6] The localized modes are referred as disorder-activated longitudinal acoustic phonon[5] and disorder-activated transverse acoustic phonon[7] modes. Furthermore, disorder-activated longitudinal optical phonon[7] and disorder-activated transverse optical phonon[7] modes have been observed recently in the Raman spectra from $Ga_{1-x}Al_xAs$.[8] However, disorder-activated modes in $Ga_{1-x}In_xAs$ have hardly been discussed up to date although those in quaternary alloy $Ga_{1-x}In_xAs_{1-y}P_y (x,y > 0)$ have been shown recently.[9]

In this letter, disorder-activated modes in Raman spectra from $Ga_{1-x}In_xAs$ epitaxial layers grown on GaAs and InP substrates and broadening of optical phonons due to stress will be reported. $Ga_{1-x}In_xAs$ epitaxial layers were grown on GaAs and InP substrates with Ga-In-$AsCl_3$-H_2 vapor phase deposition and liquid-phase epitaxy, respectively. All samples had a surface orientation of (100). Raman spectra were measured at room temperature using 514.5-nm line of Ar^+ laser as an exciting source. A power and a diameter of the laser beam were about 60 mW and about 40 μm on the surface of the sample, respectively. Raman light scattered from the sample was collected in the backscattering configuration and analyzed with a triple monochrometer JRS-400 T. Alloy compositions of the ternary alloys were determined with electron probe microanalysis.

Raman spectra from $Ga_{1-x}In_xAs$ ($x = 0 \sim 1$) epitaxial layers grown on GaAs substrates are shown in Fig. 1. Thicknesses of layers were more than 12 μm. The spectra in Fig. 1 were obtained without an analyzer. The features in the spectra denoted by *A* and *C* are LO phonons based on GaAs and InAs, respectively.[6,10] Those denoted by *B* and *D* are GaAs and InAs TO phonons, respectively.[6,10] The peak at 270 cm^{-1} corresponding to TO phonon of GaAs in the $x = 0.0$

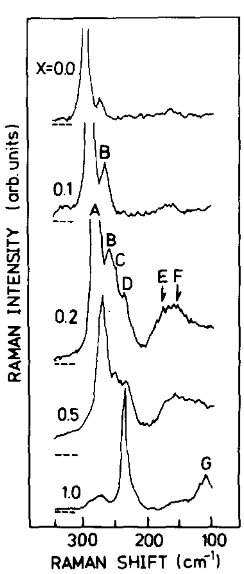

FIG. 1. Raman scattering spectra in the 100–350-cm^{-1} spectral range of $Ga_{1-x}In_xAs$ epitaxial layers of various compositions.

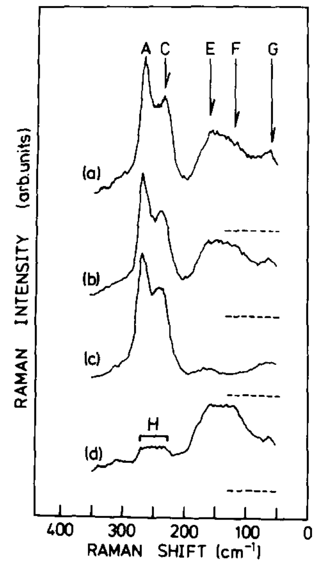

FIG. 2. Raman spectra from $Ga_{1-x}In_xAs$ ($x = 0.53$) grown on InP: (a) Unanalyzed, (b) $\Gamma_1 + \Gamma_{12} + \Gamma_{15}$ components, (c) Γ_{15} component, and (d) $\Gamma_1 + 4\Gamma_{12}$ components.

spectrum is due to the alignment of measurement slightly out of the backscattering geometry. The peak at 270 cm^{-1} in the $x = 1.0$ spectrum in Fig. 1 is based on the coupled LO phonon-plasmon mode because the carrier concentration in the InAs epitaxial layer is 1.0×10^{17} cm^{-3}. To identify other features *E*, *F*, and *G* in the spectra, Raman light was analyzed and a $Ga_{1-x}In_xAs$ ($x = 0.53$) lattice-matched layer on a InP substrate was used as a sample. Spectrum (a) obtained without using an analyzer is also shown in Fig. 2 as a reference. Features denoted by *E* and *F* in Fig. 2 are identical to those shown in Fig. 1. Peaks *B* and *D* in Fig. 1 are not evident in Fig. 2. It is considered that they are included in the tail of peak *C* because the intensity of the latter, i.e. InAs LO phonon, is strong relative to the former. Spectrum (b) in Fig. 2 was obtained in a relation in which the electric vectors of both incident and scattered lights were parallel to the {110} direction. According to the theoretical calculation by Tawler *et al.*,[11] Γ_1, Γ_{12}, and Γ_{15} components of lattice vibration are allowed to be detected in the relation. In the case of zinc-blende structure Γ_1 and Γ_{12} components are inactive theoretically. Therefore only LO phonons which correspond to Γ_{15} should appear. Peaks *E*, *F*, and *G* appear, however, in spectrum (b). Spectrum (c) was obtained in the relation that the incident and Raman lights were polarized parallel to the {100} direction and perpendicular to the incident plane, respectively. Γ_{15} component should be observed in the relation from theoretical consideration. Peaks *E* and *F* disappeared while two LO phonons *A* and *C* remained as expected. Peak *F* has been reported as 2*TA*[10] of GaAs. Peak *G* can not be identified although it is almost identical to one reported as *TA* of GaAs[10] or InAs.[7] Peak *E* at about 160 cm^{-1} is a new peak which has not been reported previously. It is considered as disorder-activated longitudinal acoustic phonon of $Ga_{1-x}In_xAs$ because its intensity is strong near the midrange of composition. Spectrum (d) in Fig. 2 was obtained in the relation that both incident and scattered lights were polarized parallel to the {100} direction. Peak *H* from about 230 to 275 cm^{-1} is also a new peak and considered to be disorder-activated optical phonon.[7] Measurements of neutron scattering are necessary to identify the new peaks strict-

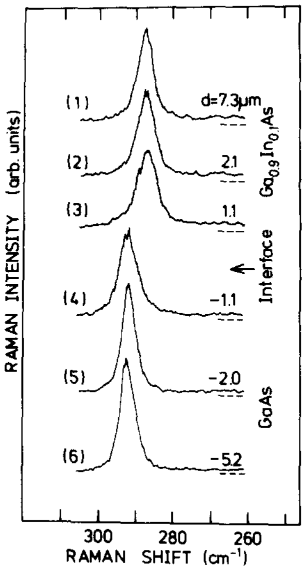

FIG. 3. Raman spectra from the regions near the interface between the $Ga_{1-x}In_xAs$ ($x = 0.1$) epitaxial layer and the GaAs substrate. *d*: distance from the interface.

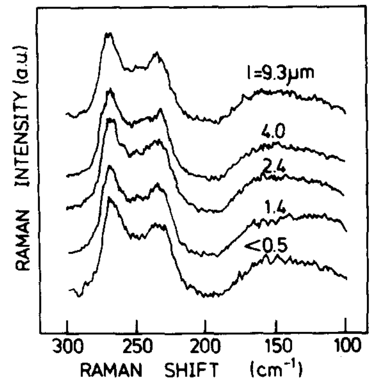

FIG. 4. Raman spectra from $Ga_{1-x}In_xAs$ ($x = 0.53$) grown on InP of various epitaxial layer thicknesses. *l*: thickness of an epitaxial layer.

ly. It can be said from the results that acoustic and disorder-activated modes are observed from $Ga_{1-x}In_xAs$ epitaxial layers in the midrange of composition independent of substrate materials when the epitaxial layers have thicknesses more than about 10 μm.

On the other hand, there is a difference in the linewidth of the LO phonon between $Ga_{1-x}In_xAs$ epitaxial layers grown on GaAs and InP substrates when the thickness of the epitaxial layers is less than about 1 μ. Raman spectra from a $Ga_{1-x}In_xAs$ ($x = 0.1$) epitaxial layer near the interface with the GaAs substrate are shown in Fig. 3. Spectra in Fig. 3 were measured by scanning a laser beam on an angle lapped and chemically etched surface of a $Ga_{1-x}In_xAs$ epitaxial layer and a GaAs substrate. The electric vectors of both incident and scattered lights were parallel to the {110} direction in the measurement. A peak at about 287 cm^{-1} is LO phonon based on GaAs and that at 292 cm^{-1} is LO phonon from GaAs substrate. The peak at 270 cm^{-1} in the $x = 0.0$ spectrum and peak *B* in the $x = 0.1$ spectrm in Fig. 1 cannot be seen in Fig. 3 because the Raman intensity drawn in Fig. 3 is lower than in Fig. 1 by about one order of magnitude while the peaks are very weak. The linewidth of the LO phonon in spectrum (3) is larger than those of the LO phonon in spectra (1) and (2) which were obtained from the epitaxial layers far from the interface. At the same time, the LO phonon in spectrum (4) which was obtained from the GaAs substrate near the interface is wider than those in spectra (5) and (6) which were measured at the substrate far from the interface. The width of the LO phonon lines from $Ga_{1-x}In_xAs$ ($x = 0.53$) lattice-matched layers on InP substrates is almost identical independent of the thicknesses of epitaxial layers as shown in Fig. 4 which shows spectra obtained from $Ga_{1-x}In_xAs$ ($x = 0.53$) epitaxial layers having different thicknesses on InP substrates. Stress in crystal has been reported as a reason for the broadening in LO phonon lines.[12] The results shown above indicate that a $Ga_{1-x}In_xAs$ ($x > 0$) epitaxial layer grown on a GaAs substrate has stress near the interface between them although lattice mismatch is only about 0.7%. The spectra in Fig. 4 were obtained without an analyzer. Features below 200 cm^{-1} in the spectra of the figure are identical independent of thicknesses of the epitaxial layers. The reason that the relative intensities among peaks *A*,*B*,*C*, and *D* are different in the $x = 0.5$ spectrum of Fig. 1 and the spectra in Fig. 4 ($x = 0.53$) has not been made clear.

In conclusion, forbidden modes appear in the Raman spectra from $Ga_{1-x}In_xAs$ epitaxial layers in the midrange of composition regardless of substrate materials and thicknesses of the layers. The modes indicate substantial existence of disorder in atomic arrangement of $Ga_{1-x}In_xAs$. On the other hand, broadening in the LO phonon line due to stress was observed from a $Ga_{1-x}In_xAs$ epitaxial layer when the layer had a thickness less than 1 μ and lattice mismatch with substrate.

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