

Raman spectra of GaAs with ultrathin InAs layers inserted

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Raman spectra of GaAs with ultrathin InAs layers inserted are measured and interpreted by comparing with spectra calculated based on the linear-chain model. The samples are grown by molecular-beam epitaxy, and the InAs layer thickness is varied from 2 ML (monolayers) to 10 ML. Both (001)-oriented and slightly misoriented substrates are used. From the transmission electron micrograph observation, it is found that the introduction of misfit dislocations is delayed on the misoriented substrate compared with the oriented substrate. When the InAs thickness is less than 6 ML, optical phonon modes in InAs are not observed. This is due to coupling of the InAs vibration to the vibration of the GaAs host lattice. At a thickness of 6 ML, the InAs longitudinal-optical mode begins to appear for the sample on the oriented substrate, but it is still unobservable for that on the misoriented substrate. Thus, the intensity of the InAs mode is stronger in a more heavily dislocated structure. This is because the InAs vibration is decoupled from the host GaAs vibration owing to dislocations at the heterointerfaces.

I. INTRODUCTION

A substitutional impurity atom in a crystal may give rise to localized modes and/or resonance modes of oscillation, and the conditions for these different types of behaviors are now well understood.¹ An In atom in GaAs has been shown to exhibit the resonance-type behavior, and this fact can be understood by the following consideration: Since the In atom is heavier than the atom to be substituted for (i.e., the Ga atom), it is obvious that the In atom does not have a localized mode with frequency above the optical branch of GaAs. In addition, since GaAs has a very narrow frequency gap because of similarity of masses of Ga and As, it is not probable that GaAs has a gap mode. In other words, the phonon bands of InAs overlap completely with the phonon bands of GaAs. Thus an In atom gives rise to resonance modes, i.e., the In-As vibration couples with the Ga-As vibration.

Although the behaviors of the isolated impurities have been thus clarified, it has scarcely been investigated what happens if the impurity atoms nucleate to form a two-dimensional layer or a three-dimensional cluster. For a layer or cluster of a sufficiently large size, the phonon mode localized in it will be observed, even if the localized mode is not observable for the isolated atom. Then, the important question is how large the size should be for the localized mode to be observable. However, if there is a lattice mismatch between the host crystal and the impurity layer or cluster, misfit dislocations are necessarily introduced at the boundary for a layer or cluster larger than a certain critical size. Thus another important question is how the dislocations influence the behavior of the impurity mode.

In this study, we characterize ultrathin InAs layers embedded in GaAs by the Raman spectroscopy. The Ra-

man spectroscopy has a sensitivity of the order of a few atomic layers, if the excitation light is properly chosen, and thus it has been regarded as a powerful tool for characterizing thin semiconductor layers. So far, Raman spectra of ultrathin embedded layers have been reported for Ge in Si (Ref. 2) and Si in GaAs (Ref. 3). However, a Ge atom in Si and a Si atom in GaAs both cause localized lattice vibrations. Thus there is an important difference between the InAs/GaAs system and the Ge/Si or Si/GaAs system. For the latter systems, one can predict from the present understanding on the impurity mode that the phonon mode of the impurity layer is in principle observable even for a single atomic layer. On the other hand, it may depend on the layer thickness or the dislocation density whether the phonon in the InAs layer embedded in GaAs is observable or not.

Aside from the interest in the lattice dynamics, the growth of InAs on GaAs has been of considerable interest in the semiconductor growth technology. The lattice mismatch between GaAs and InAs is very large, about 7%, and such a large mismatch strain is expected to cause the growth mode change and defect generation in the heteroepitaxial growth. Thus the initial stage of the growth and the crystalline structure of the interface have been extensively studied. In molecular-beam epitaxial (MBE) growth, the initial stage can be observed *in situ* by reflection high-energy electron diffraction (RHEED). Reported results of RHEED studies show that InAs layers on (001) GaAs remain coherent, i.e., fully strained for thicknesses up to 0.5 nm or about 2 ML (monolayers).⁴ Relaxation becomes significant when the thickness exceeds this value. On the other hand, three-dimensional (3D) islands begin to form at nearly the same thickness.

Some tried to control the initial stage of the InAs growth on GaAs. Grandjean, Massies, and Etgens used Te as a surfactant in MBE growth of InAs/GaAs and

found that the 2D and coherent growth of InAs is sustained up to 6 ML with a Te surfactant layer.⁵ On the other hand, Tabuchi and Sasaki found that the misfit dislocation generation is delayed by using misoriented substrates.⁶ They explained the results considering the interaction of the dislocation with the steps on the substrate surface. Similar results were reported recently for $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ by Maigne and Roth.⁷

The optical properties of InAs/GaAs have also been extensively studied. Recently, it was found that GaAs with inserted ultrathin InAs layers exhibits intense photoluminescence associated with the inserted InAs layers.⁸ However, the luminescence properties are degraded by dislocations since dislocations usually act as a nonradiative recombination center. Thus the crystalline structure is of great interest from this viewpoint, too.

In the next section, we describe properties of samples used in this study. Our samples are MBE grown GaAs/InAs/GaAs structures with InAs layer thicknesses varied from 2 to 10 ML. Misoriented GaAs substrates are used in addition to (001)-oriented ones. As described above, the dislocation generation is delayed on the misoriented substrate, and thus we can investigate the influence of dislocations on phonon properties by comparing two samples with the same InAs thickness but on different substrates. The results of the Raman experiments are given in Sec. III. In Sec. IV we carry out a theoretical calculation based on the linear-chain model to interpret observed Raman spectra. Although the calculation is based on many simplifying assumptions, it succeeds in explaining some features of the observed spectra.

II. SAMPLES

The GaAs substrates used in this study are nominally (001) ($\pm 0.5^\circ$) and 3.5° misoriented toward $[1\bar{1}0]$. On these substrates, a 100-nm GaAs buffer layer, a thin InAs layer, and a 10-nm GaAs cap layer were grown successively. The InAs layer thickness is either 2, 4, 6, 8, or 10 ML. Substrate temperature was kept at 590°C during the growth of the buffer layer and then lowered to 480°C for the growth of the InAs layers and the cap layers. The two kinds of substrates were set on the same molybdenum block in the MBE chamber so that the layer thickness grown is equal for both the substrates.

RHEED observation during the growth shows that the island formation starts at a thickness of about 1.5 ML on both types of substrate. Thus the InAs layers in the present samples are not flat 2D layers, and the value of the InAs thickness is an average one.

Dislocations in the grown samples were observed by plan-view transmission electron micrograph (TEM). No dislocation was observed when the InAs thickness is 2 ML. When the thickness is 4 ML, misfit dislocations composed of a couple of 60° -type dislocations were found to run along $[1\bar{1}0]$ in the heterostructure on the oriented substrate. On the other hand, dislocations are still absent for the thickness of 4 ML on the misoriented substrate. When the InAs thickness is increased to 6 ML, the misfit

dislocations were found in the structure on the misoriented substrate, too. However, the dislocation density is smaller by a factor of 2 than on the oriented substrate. When the InAs thickness is larger than 7 ML, the structures grown were heavily dislocated even on the misoriented substrate.

It has been reported that In atoms are spread into upper GaAs layers because of the surface segregation during the MBE growth of GaAs on InAs. For example, Moison *et al.* reported that about 80% of a deposited single monolayer of InAs is spread into the GaAs overlayer.⁹ Similar segregation may occur in the growth of the present samples, although we have not actually measured the degree of mixing. Therefore the actual InAs layer thickness after the growth of the final cap layer will be smaller than that of the originally deposited InAs layer, and the cap layer will become an $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloy rather than pure GaAs, especially near the interface. Influence of the segregation on the Raman properties is qualitatively discussed in the later section.

III. RAMAN SPECTRA

Raman Spectra were taken at room temperature using the 488-nm line of an Ar laser as a light source in a quasi-backscattering geometry. The polarization vector of the incident light is parallel to $\langle 100 \rangle$ of the substrate. The scattered light is analyzed by a double monochromator and detected by a photomultiplier tube. The spectral resolution is 5 cm^{-1} .

Electron-hole pairs are created by the laser beam during the Raman measurement, and one might expect that the excited carriers accumulate near the InAs layer and give rise to phonon-plasmon coupled modes. However, since the cap GaAs layer is thin (10 nm), the InAs layer is included in the surface depletion region and the carrier concentration will not be very high there. Thus we believe that the effect of the carrier is not significant.

Raman signals associated with the ultrathin InAs layer are expected to be very weak compared with the signal due to the GaAs longitudinal-optical (LO) phonon. In addition, the second-order Raman signal of GaAs and weak plasma lines of the Ar laser may disturb the spectra. Thus we first measured the spectrum for pure GaAs and subtracted it from the spectra of the GaAs/InAs/GaAs structures. Before the subtraction, we normalized the intensity of the spectra so that the intensity of the GaAs LO peak becomes equal. The difference spectrum thus obtained in general includes the contribution of perturbed GaAs vibrations as well as that of InAs vibrations. In fact, we observed an enhanced tailing of the GaAs LO peak as described below.

Figure 1 shows examples of the measured Raman spectra; the upper spectrum is for the structure with 4ML InAs on the oriented substrate, and the lower spectrum for pure GaAs. Figure 2 shows spectra obtained by the subtraction procedure. The configuration $z(xy)\bar{z}$ corresponds to the cross polarization of the incident and scattered light, and the configuration $z(xx)\bar{z}$ to the parallel polarization. The scattering by the LO mode in a zincblende crystal is allowed in $z(xy)\bar{z}$. No structure is ob-

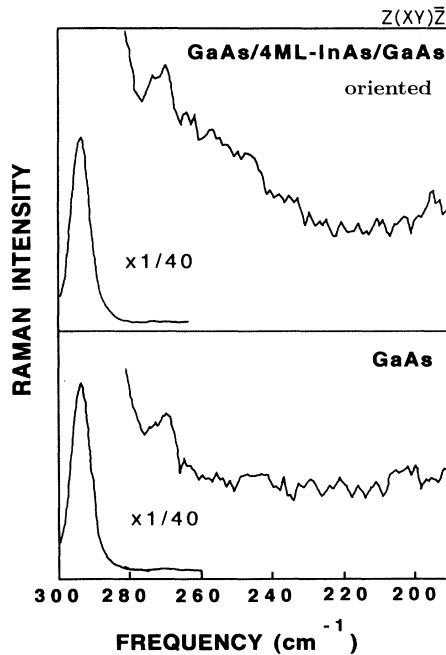


FIG. 1. $z(xy)\bar{z}$ Raman spectra of pure GaAs and GaAs with 4-ML-thick InAs inserted.

served except a sharp peak at 270 cm^{-1} in the spectra in Fig. 2(a): The Raman spectra of the samples with 2 ML InAs are not appreciably different from that for pure GaAs. The peak at 270 cm^{-1} is due to the transverse-optical (TO) phonon in GaAs. Although the scattering by the TO mode is forbidden in a backscattering configuration on a (001) face, it is observed because of deviation in the configuration and/or misorientation of the substrate. As shown in the figure, the peak is observed more strongly for the misoriented substrates than for the oriented ones.

The effects of the inserted InAs layers on the Raman spectra are clearly seen for the structures with 4 ML InAs. As shown in Fig. 2(b), the signal intensity of the $z(xy)\bar{z}$ spectra gradually increases with frequency in the region above 230 cm^{-1} . A hump is seen near 260 cm^{-1} in the $z(xy)\bar{z}$ spectrum of only the oriented sample. An increase in the intensity above 260 cm^{-1} is also seen in the $z(xx)\bar{z}$ spectrum for the oriented sample, but it is not clearly observed for the misoriented one.

In Figs. 2(a) and 2(b), a peak is not observed near the frequency of the zone-center LO mode in InAs, i.e., 241 cm^{-1} . For the heterostructure with 6 ML InAs on the oriented substrate, a broad peak appears near 240 cm^{-1} in the $z(xy)\bar{z}$ spectrum, as shown in Fig. 2(c). A very broad peak is also observed in the $z(xx)\bar{z}$ configuration, in which the scattering by the LO mode is forbidden. The gradual increase in the signal intensity toward higher frequency appears as in Fig. 2(b), and the broad peak overlaps this gradual increase. On the other hand, the peak is not observed for the misoriented sample, although the gradual increase in the $z(xy)\bar{z}$ spectrum becomes larger than in Fig. 2(b). No signal is detected in $z(xx)\bar{z}$

except for the GaAs TO peak, which appears mainly because of the misorientation.

Figure 2(d) shows spectra for the samples with 8 ML InAs. The vertical scale of this figure is contracted by a factor of 2 compared with Figs. 2(a)–2(c). Thus the intensity of the peak near 240 cm^{-1} for the oriented samples increases by a factor of about 2 as the InAs layer thickness is increased from 6 to 8 ML. In the $z(xx)\bar{z}$ spectrum, a weak peak appears near 220 cm^{-1} , the frequency of the zone-center TO mode in InAs. The spectra for the misoriented sample are not qualitatively different from the oriented one, although the intensity is slightly smaller for the misoriented sample.

Spectra for the structures with 10 ML InAs are similar to those in Fig. 2(d) but much more intense. The $z(xy)\bar{z}$ spectrum intensity increases by a factor of 3 and the $z(xx)\bar{z}$ spectrum intensity by a factor of 4. In addition, the peak near 220 cm^{-1} becomes sharp.

IV. MODEL CALCULATION

A. Calculation procedure

Before interpreting the experimental results, we give the spectra calculated using the linear-chain model. This model has proven to be useful in interpreting Raman spectra of semiconductor superlattices.^{10,11} However, one should not expect quantitative agreement between the experimental results and the calculated results given below because the calculation is based on many simplifying assumptions. Nevertheless, the calculation is of great use in discussion in the next section.

To simulate the samples characterized in this study, we consider a linear chain composed of 35 pairs of Ga and As atoms (35 ML of GaAs), n pairs of In and As atoms, and 80 pairs of Ga and As atoms. The first part corresponds to the 10-nm-thick GaAs cap layer, and the third part is considered to simulate the GaAs buffer layer and the substrate. (It was confirmed that the calculated Raman spectrum of the 80-ML-long GaAs chain is little different from that of the ideal infinite crystal.) Both the ends of the chain are an As atom, and thus the total number of atoms in the chain, N , is $2(35 + n + 80) + 1$.

Frequencies and eigenvectors of vibration of the chain are obtained by diagonalizing the $N \times N$ matrix which describes the motion of the N atom chain.

We take account of only the nearest-neighbor interaction, and the force constant is chosen so that the zone-center optical frequency of the infinite Ga-As (In-As) chain becomes equal to the LO frequency of GaAs (InAs). Change of the force constant due to the misfit strain is not taken into account. In addition, the long-range interactions, including the Coulomb interaction, are neglected. The ends of the chain are set free.

For comparison, vibrations of the following two types of chains are also calculated. The first one is n monolayers of InAs with free-surface boundaries. The second one is an InAs/GaAs/InAs structure, i.e., InAs with an ultrathin GaAs layer inserted.

The Raman scattering intensity of each vibration mode is calculated on the basis of the bond polarization model.

In the linear chain, each atom is thought to represent one (001) atom plane, and each bond to represent two-dimensional array of bonds on a (001) plane. In a zinc-blende lattice, direction of bonds on a (001) plane is different from direction of bonds on the next plane. Thus two consecutive bonds in the linear chain are thought to have different polarizability since they represent bonds of

different directions. The LO scattering on a (001) plane is caused by this polarizability difference. The $z(xy)\bar{z}$ scattering intensity of each vibration mode is given by¹¹

$$I_{xy} \propto \left[\sum \{ (u_{2n+1} - u_{2n}) - (u_{2n} - u_{2n-1}) \} \right]^2 \\ = 4 \left[\sum (u_{2n} - u_{2n-1}) \right]^2,$$

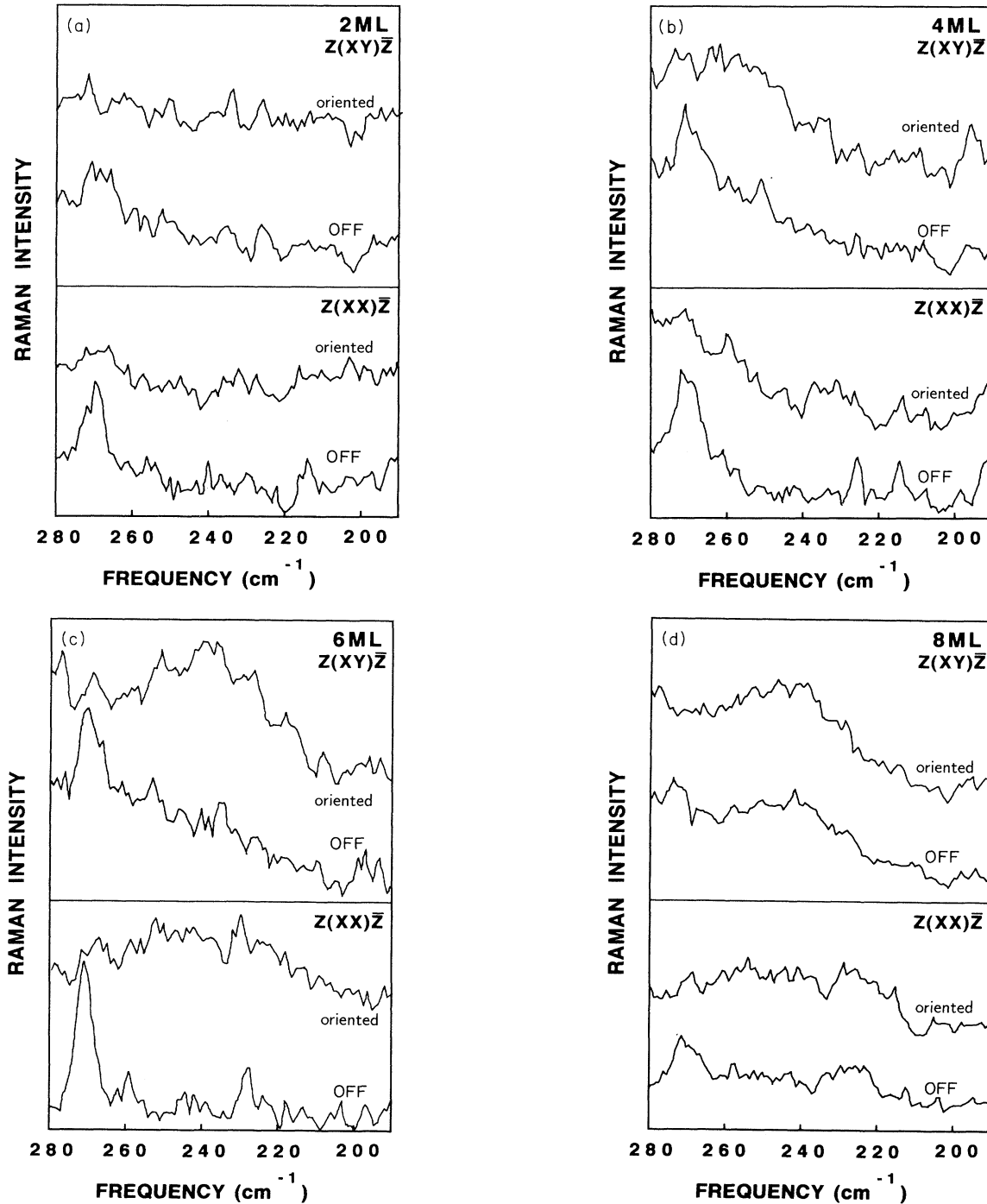


FIG. 2. Raman spectra of GaAs/InAs/GaAs structures with different InAs layer thicknesses after subtraction of the spectrum of pure GaAs. (a) 2 ML, (b) 4 ML, (c) 6 ML, and (d) 8 ML. The spectra labeled "oriented" are for the samples on the (001) substrates, and those labeled "OFF" are for the samples on the misoriented substrates.

where u_i is the displacement of the i th atom. For simplicity, difference in polarizability between InAs and GaAs is neglected, and the wavelength of the incident light is assumed to be infinite.

Raman spectra are generated by broadening each vibration peak to take account of the natural linewidth and the instrumental resolution, and superimposing them after multiplying their intensity by the Bose factor.

B. Calculation results

Figure 3 shows examples of the calculated Raman spectra; the bold solid line represents spectrum for $n=2$ (GaAs/2 ML-InAs/GaAs) and the thin solid line that for $n=0$ (pure GaAs). We calculate the difference between the spectra for the pure GaAs chain and for the GaAs/InAs/GaAs chain, as we did for the experimental spectra. Figure 4 shows the difference spectra. For comparison, spectra for the free InAs chains are also shown by thin solid lines.

When $n=2$, a well-defined InAs phonon peak is not observed for the GaAs/InAs/GaAs structure. For $n=4$, the InAs peak is clearly observed but is still broad. With increase in n from 4 to 6, its peak height increases by a factor of 2. Small increase in the Raman signal compared with pure GaAs is observed in a wide frequency range above 250 cm^{-1} . This shows that the tailing of the GaAs peak toward lower frequency is enhanced in the GaAs/InAs/GaAs structures. The GaAs peak slightly shifts toward lower frequency in the GaAs/InAs/GaAs structure, and thus the intensity of the difference spectra is negative at frequencies above 293 cm^{-1} .

On the other hand, the calculated Raman spectra for the free InAs chains consist of discrete peaks, and the frequency of the highest-frequency peak approaches 241 cm^{-1} as n increases. It should be noted that although 2-ML-thick InAs exhibits a sharp peak at 230 cm^{-1} under the free-boundary condition, it exhibits only a broad signal near that frequency when sandwiched by GaAs.

The InAs peaks of the GaAs/InAs/GaAs structures (the peak in the range $230\text{--}240 \text{ cm}^{-1}$) do not correspond to a single vibration mode but are composed of peaks of several different modes. An analysis of the eigenvector of

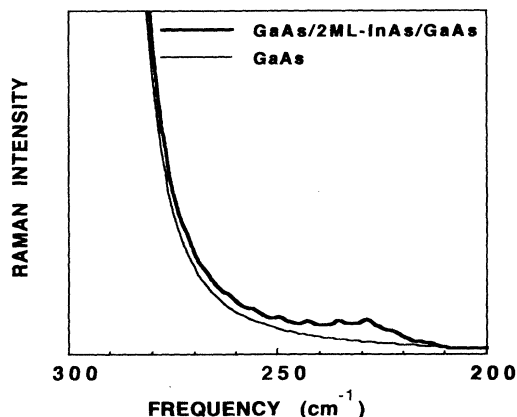


FIG. 3. Calculated $z(xy)\bar{z}$ Raman spectra for the GaAs chain and the GaAs/2 ML-InAs/GaAs chain.

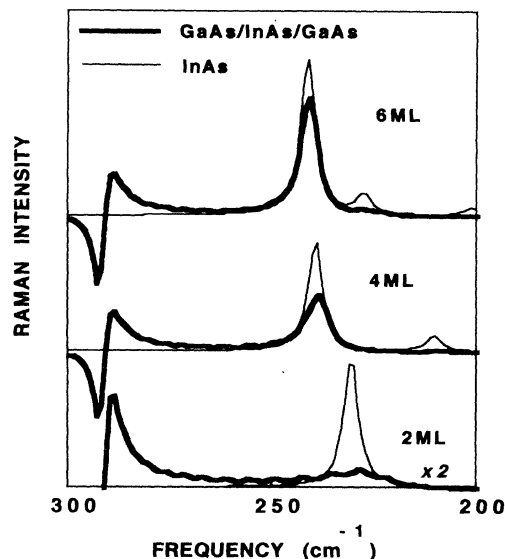


FIG. 4. Calculated $z(xy)\bar{z}$ Raman spectra for the GaAs/ n ML-InAs/GaAs chains after subtraction of the spectrum of the pure GaAs chain (the bold lines). Because of the subtraction, the intensity is negative at some frequencies. For comparison, spectra for the n -ML InAs chains with a free boundary are also shown by the thin solid lines.

these modes shows that they have significant vibration amplitude not only in the InAs layer but also in the GaAs layers, i.e., the InAs optical vibrations are coupled to the GaAs vibrations. The GaAs vibrations coupled with the InAs vibrations are of an optical branch near the Brillouin-zone edge, and some of them are localized in the cap part, some are localized in the substrate part, and the others propagate through the whole chain. The frequency of the "InAs" mode is different when it is coupled to a different GaAs mode. Thus the peak becomes broad and even unobservable for $n=2$. As the InAs thickness increases, the modes are grouped more distinctly into modes localized in the GaAs parts and those in the InAs part.

The enhanced tailing of the GaAs peak toward lower frequency is due to the localization of the GaAs mode, especially in the rather thin cap part: the localization increases the scattering intensity of the mode with a finite wave number, which has null scattering efficiency in the infinite chain.

In the calculated Raman spectra of the InAs/ n ML-GaAs/InAs, the GaAs peak clearly appears even for $n=2$. This can be easily understood by the fact that the optical mode of GaAs has higher frequency than any modes in InAs and thus is hardly coupled to them.

V. DISCUSSION

A. Dependence on the InAs layer thickness

In the experimental results, the Raman signal attributed to the vibration in InAs is absent when the InAs thickness is small: it becomes observable when the thickness is 6 ML on the oriented substrate and 8 ML on the misoriented substrate. Once the peak appears, its intensi-

ty increases superlinearly with the InAs thickness. The same tendency is also seen in the calculated spectra and can be explained considering the coupling of the InAs mode with the GaAs mode as discussed in Sec. IV. Therefore we believe that the coupling makes the InAs peak unobservable for the samples with very thin InAs layers.

As shown in Fig. 2, the gradual increase in the Raman signal with frequency is detected in the range above 250 cm^{-1} even when the InAs peak is not observed. This signal increase will be regarded as tailing of the GaAs LO peak. The enhancement of the tailing is also reproduced by the calculation and can be attributed to the localization of the GaAs phonon in the cap layer. The spreading of In atoms into the upper GaAs layer may also contribute to the down shift of the peak and the enhanced tailing in the lower-frequency side. A down shift of the peak may also be caused by a tensile strain. When the InAs layer is relaxed, the GaAs layer grown on it is tensilely strained. However, since the top GaAs layer is 10 nm thick, most of the misfit strain will be relaxed by dislocations at the upper GaAs/InAs interface. Therefore the effect of the residual strain in the top GaAs layer will not be significant.

Although qualitative tendencies of the calculated spectra agree with those of the experimental ones, there are some quantitative differences between them. The InAs peak clearly appears in the calculated spectrum for $n = 4$ but not in the experimental spectra for the samples with 4 ML InAs. In addition, the observed InAs peaks are much broader than the calculated ones. Obviously, one reason for these discrepancies will be the fact that many assumptions are used in the calculation. In addition, the samples are in fact not an ideal layered structure but are disturbed by the island formation and the segregation. In particular, the segregation will influence the dependence of the spectra on the InAs thickness. The actual thickness of the InAs layer embedded in GaAs will be smaller than the nominal thickness because of the loss of In atoms by the segregation. This may partly explain the fact that the InAs peak begins to appear at a larger InAs thickness than predicted by the calculation.

A hump appears near 260 cm^{-1} in the $z(xy)\bar{z}$ spectrum for the oriented samples with 4 ML InAs. We have not clarified the origin of the hump. It may correspond to the InAs LO mode shifted by misfit strain; if the InAs layer is coherent to GaAs, the peak will be shifted to about 265 cm^{-1} .¹² Since the strain has already been relaxed to some extent in this sample, it is possible to assign the hump to the LO mode in partly relaxed InAs. However, it will also be possible to attribute the hump to some structures of the GaAs dispersion curves and relaxation of the momentum conservation rule.

Effects of the island formation are not understood. To discuss the effects, we should use a three-dimensional model of the lattice vibration instead of the linear-chain model.

B. Influence of dislocations

As shown in Fig. 2, significant differences in Raman spectra are found between the GaAs/InAs/GaAs struc-

tures on the oriented substrates and those on the misoriented substrate. For example, the InAs peak is observed for the structure with 6 ML InAs on the oriented substrate but not on the misoriented substrate. Since the InAs thickness is the same for both cases, the differences will be due to the difference in the dislocation density: As described in Sec. II, the dislocation density is larger in the samples on the oriented substrates.

Lattice defects such as dislocation are expected to interrupt the propagation of phonons. Thus the phonons in InAs and those in GaAs will be coupled less strongly when a larger number of dislocations are introduced at the interface between them. According to the calculated results shown in Fig. 4, the InAs peak height is larger for the InAs layer with free boundaries than for the embedded layer of the same thickness, i.e., the InAs peak height is increased by the decoupling from the GaAs layers. Therefore the propagation interruption by dislocations is expected to enhance the Raman peak of the InAs mode. This will explain the result that the InAs peak is observed more clearly for the more heavily dislocated samples, i.e., the samples on the oriented substrates. When the nominal InAs layer thickness is equal to or larger than 8 ML, the structure on the misoriented substrate is also heavily dislocated and thus the difference due to the orientation is not significant.

The dislocations influence other features of the spectra. The $z(xx)\bar{z}$ spectrum intensity tends to be larger for more heavily dislocated samples: it increases with the InAs thickness and is larger for the oriented samples than for the misoriented ones, as shown in Fig. 2. The $z(xx)\bar{z}$ scattering intensity predicted from the calculation is negligibly small in the optical phonon frequency range, and thus the observed $z(xx)\bar{z}$ scattering is believed to be activated by some structural disorder. Therefore the $z(xx)\bar{z}$ intensity is thought to be the measure of the misfit dislocation density in the GaAs/InAs/GaAs structures. The InAs TO phonon scattering observed for the samples with 8 ML InAs will also be activated by the dislocations.

The observed InAs peaks are very broad, broader than the predicted ones. This may be partly due to the inhomogeneous relaxation of the misfit strain. The mismatch between InAs and GaAs is about 7%, and strain of 7% causes a shift of about 24 cm^{-1} in the InAs LO frequency. The misfit strain is compensated by the strain due to the misfit dislocation. Since the strain field of a dislocation is not uniform, there should be microscopic variation in the amount of strain near the dislocation. Thus the Raman peak broadens for the ultrathin dislocated layers.

C. Comparison with other structures

Brodsky and Lucovsky studied behavior of optical phonons in $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloys by use of infrared reflectivity measurements.¹³ Their results show that $\text{In}_x\text{Ga}_{1-x}\text{As}$ exhibits the partially two-mode behavior, i.e., the InAs mode is observed only when the In content exceeds a threshold value ($x = 0.2$). This fact can be compared to the present result that the optical phonon in the embedded InAs layer is observed only when its thickness exceeds a critical value. In a random alloy, the aver-

age size of InAs clusters increases with increasing InAs content. The InAs mode will become observable when sufficiently large InAs clusters are formed in a significant number, and thus the InAs mode in the alloy is observed only for sufficiently large x . However, it will be difficult to estimate the critical cluster size or the threshold composition from the present results; the sample of the present study is a layered structure, while the alloy has three-dimensional randomness of atom arrangement.

Gerard *et al.* reported Raman spectra of an $(\text{InAs})_4(\text{GaAs})_3$ superlattice.¹⁴ Three peaks appear in the optical phonon frequency range in the $z(xy)\bar{z}$ spectrum, and two of them are attributed to the LO modes confined in the GaAs layers and the other is attributed to a propagative mode mostly of InAs character. On the other hand, we do not observe the InAs-like mode for the single embedded layer of a thickness of 4 ML. We think that the repetition of many layers makes the InAs mode observable in the superlattice.

We calculated the Raman spectra of the $(\text{InAs})_m(\text{GaAs})_m$ superlattices using the model described in Sec. IV. The calculated spectrum for $m=4$ agrees qualitatively with the spectrum in Ref. 14: two localized modes and one propagative mode appear in the frequency range between 230 and 290 cm^{-1} . A sharp peak attributed to the InAs-like propagative mode appears in the spectrum for $m=2$, although the InAs modes form a broad structure for the single inserted 2-ML-thick InAs layer as shown in Fig. 4. This shows that the InAs mode can be observed more clearly for a periodic structure than for a single embedded layer. On the other hand, the $(\text{InAs})_1(\text{GaAs})_1$ superlattice exhibits only one strong peak in the range above 200 cm^{-1} , according to the calculation results. Its peak frequency is about 270 cm^{-1} , an intermediate value between GaAs LO and InAs LO fre-

quencies, and the vibrational amplitudes of Ga and In atoms in this mode are comparable. Thus the one-mode behavior is ascribed to the $(\text{InAs})_1(\text{GaAs})_1$ superlattice. This is in contrast to the fact that the two-mode behavior is ascribed to both the superlattices with $m \geq 2$ and the random alloy of composition $x=0.5$. This indicates that the InAs-like phonon becomes observable only when the size of InAs clusters or the thickness of InAs layers becomes larger than a certain critical value.

VI. SUMMARY

Raman spectra of GaAs with ultrathin InAs layers inserted have been measured. The structures were grown on both (001)-oriented and slightly misoriented substrates. The misorientation delays the introduction of misfit dislocations. When the InAs thickness is less than 6 ML, optical phonon modes in InAs are not observed, probably because of coupling of the InAs vibration to the vibration of the GaAs host lattice. At a thickness of 6 ML, the InAs longitudinal-optical mode begins to be observed for the sample on the oriented substrate but not for that on the misoriented substrate. Thus the intensity of the InAs mode is stronger in a more heavily dislocated structure. This will be due to decoupling of the InAs vibration from the host vibration by dislocations.

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