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Raman study of self-assembled GaAs and AlAs islands embedded in InAs

D. A. Tenne*
Institute of Semiconductor Physics, pr. Lavrenteva 13, 630090 Novosibirsk, Russia
and Institut für Physik, Technische Universität Chemnitz, D-09107 Chemnitz, Germany

V. A. Haisler, A. I. Toropov, A. K. Bakarov, and A. K. Gutakovsky
Institute of Semiconductor Physics, pr. Lavrenteva 13, 630090 Novosibirsk, Russia

D. R. T. Zahn
Institut für Physik, Technische Universität Chemnitz, D-09107 Chemnitz, Germany

A. P. Shebanin
United Institute of Geology, Geophysics and Mineralogy, pr. Koptjuga 3, 630090 Novosibirsk, Russia

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Vibrational spectra of self-assembled GaAs and AlAs islands grown by molecular-beam epitaxy on InAs (001) substrates have been investigated by Raman spectroscopy. We observed large strain-induced shifts of optical-phonon frequencies in GaAs and AlAs islands with respect to the bulk materials. The values of shifts are 36 and 24 cm^{-1} for GaAs longitudinal-optical (LO) and transverse-optical (TO) phonons and 55 and 28 cm^{-1} for AlAs LO and TO phonons, respectively. Comparison of experimental data with calculated optical-phonon frequencies in strained GaAs and AlAs islands of different shape demonstrates that the structures studied are coherently strained, i.e., they do not contain dislocations that could lead to strain relaxation. The features of interface phonons were observed in the polarized Raman spectra between the peaks of InAs TO and LO phonons. The frequency positions of interface phonon lines are well described by the dielectric continuum model and also give evidence of three-dimensional island formation. Doublets of folded acoustic phonons appear in the low-frequency region of Raman spectra of multilayer structures with GaAs and AlAs islands. These doublets are very similar to those typically observed in planar superlattices and well described by the elastic continuum model.

I. INTRODUCTION

Semiconductor quantum dots (QD's) attract much research interest due to a range of new physical phenomena caused by three-dimensional confinement of carriers. On the other hand, zero-dimensional semiconductor structures have promising properties for new device applications such as lasers with improved performance.[1,2] Self-assembling of nanostructures during heteroepitaxy[2,3] is considered as one of the most advantageous ways of *in situ* QD fabrication without processing by lithography and etching. It is well known that the deposition of a material having large lattice mismatch with respect to the substrate under certain conditions leads to spontaneous formation of three-dimensional islands (Stranski-Krastanov growth mode). The most intensively investigated material system is InAs QD's on a GaAs substrate, which became a model system for such a type of nanostructures. Up to now, most studies have dealt with the mechanisms of island formation, their structure, and electronic and optical properties (for a review see, e.g., Ref. 2). Vibrational spectra of self-assembled QD's are far less investigated. For InAs dots in a GaAs matrix, there are data about optical phonons obtained by resonant photoluminescence[2,4] and theoretical calculations of optical-phonon energies taking into account strain distribution in QD's.[5] Recently, Pusep *et al.*[6] reported the Raman study of interface phonons in this system. Several groups also reported the phonon Raman scattering studies of QD's grown in other heteroepitaxial systems [Ge/Si,[7,8] (In,Ga,Al)Sb/GaAs,[9] InSb/InP,[10] and InAs/InP (Refs. 11 and 12)].

In this paper we present an experimental study of self-assembled GaAs and AlAs nanometer-sized clusters in an InAs matrix. The phonon spectra of these nanostructures in the whole frequency range have been investigated by Raman spectroscopy. Since the band gaps of GaAs and AlAs are larger than that of InAs, these clusters have no confined electronic or hole states. Such structures are sometimes called ''antidots.'' However, optical phonons are confined inside GaAs (AlAs) clusters in the same manner as phonons of InAs QD's in a GaAs (AlAs) matrix. So, GaAs and AlAs islands embedded in InAs can be considered as ''phonon dots.'' A specific feature of this system compared to other strained heteroepitaxial systems studied before [InAs on GaAs substrate,[2–6] Ge on Si,[7,8,13] (In,Ga,Al)Sb on GaAs,[9] CdSe on ZnSe (Ref. 14)] is a smaller lattice constant of the cluster material with respect to the substrate. This determines the opposite sign of strain (clusters are stretched). To the best of our knowledge, self-assembled tensile-strained GaAs and AlAs nanoclusters have not been investigated so far. (There were resonant Raman scattering results attributed to the possible cluster formation in $\mathrm{In}_x\mathrm{Al}_{1-x}\mathrm{As}$ alloys.[15] However, those structures were lattice matched to InP substrates.)

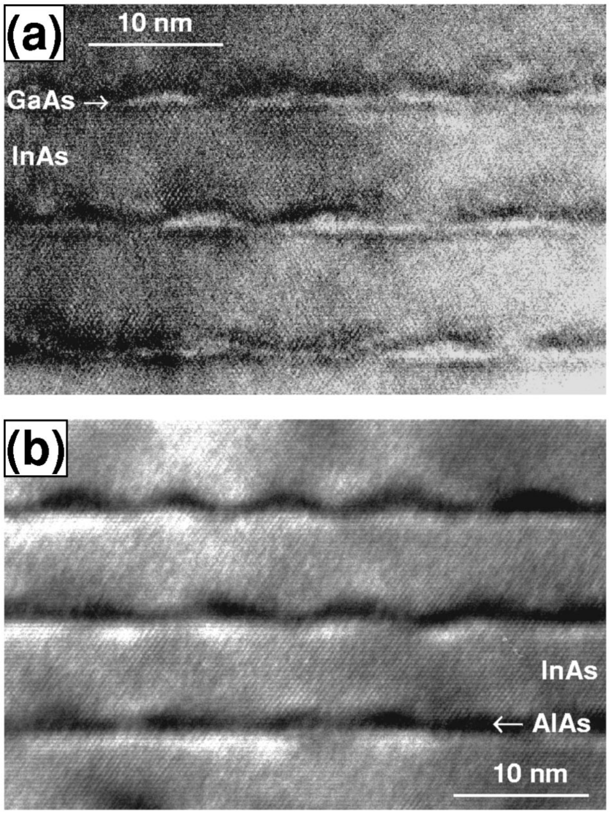

FIG. 1. High-resolution cross-sectional TEM images of samples *C* (a) and *D* (b).

II. EXPERIMENT

The structures with GaAs and AlAs clusters were grown by molecular-beam epitaxy on InAs (001) oriented substrates at temperatures in the range of 430–445 °C. Deposition rates of GaAs, AlAs, and InAs were 0.06, 0.04, and 0.25 monolayers per second, respectively. The growth was monitored by reflection high-energy electron diffraction (RHEED). According to RHEED data, the transition from a two-dimensional to a three-dimensional growth mode (beginning of island formation) occurs after the deposition of 1.8 monolayers of GaAs or AlAs. Multilayer structures with GaAs and AlAs clusters were grown in order to increase the Raman signal from the clusters. The samples studied consist of five layers of GaAs or AlAs clusters separated by InAs spacer layers. The spacer thickness is 10 nm for samples *A*,*B* and 8 nm for samples *C*,*D*. The amount of deposited GaAs was equal to 5, 3.5, and 2 monolayers for samples *A*,*B*, and *C*, respectively. Sample *D* is completely analogous to sample *C*, but contains AlAs clusters instead of GaAs. After the deposition of the nominal amount of GaAs (AlAs), the growth was interrupted for 45–50 sec before the growth of the InAs spacer allowing islands to reach equilibrium sizes. According to RHEED data, the surface becomes smooth again after the growth of 4–5 monolayers of an InAs spacer. The top layer of GaAs (AlAs) islands was covered by a 2-nm InAs cap layer.

Figure 1 shows cross-sectional images of the samples *C* and *D* obtained by high-resolution transmission electron microscopy (TEM). The average sizes of AlAs islands determined by TEM are 4–5-nm base length and 1.5–2-nm height (in the growth direction). According to TEM data, GaAs islands have larger base lengths (6–7 nm) while the height is approximately the same (about 1.5 nm).

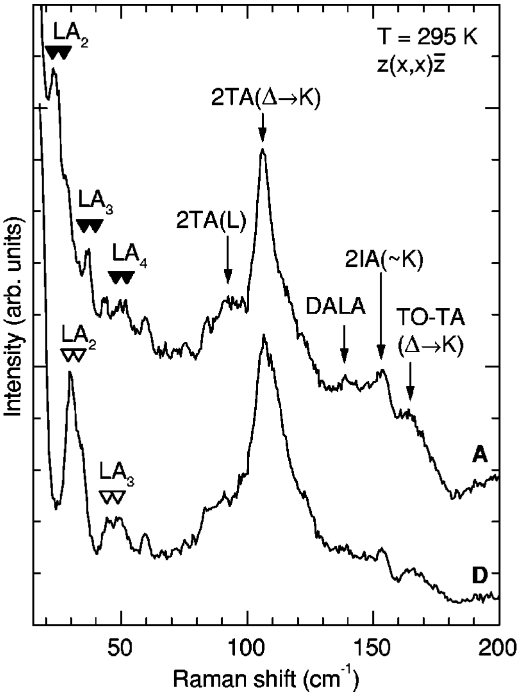

FIG. 2. Low-frequency Raman spectra of samples *A* and *D*. Triangles show the positions of folded LA doublets calculated using the elastic continuum model.

Raman spectra were recorded at 80 and 300 K in the backscattering geometry. We used a Dilor *XY* triple spectrometer equipped with a multichannel CCD detector and Jobin Yvon U1000 double spectrometer with GaAs photomultiplier. Raman scattering was excited by the 514.5-nm line of an Ar$^+$ laser.

III. RESULTS AND DISCUSSION

A. Low-frequency region

Figure 2 shows the Raman spectra of samples *A* and *D* in the low-frequency region. The spectra contain the doublets of folded longitudinal-acoustic (LA) phonons similar to those of periodic two-dimensional structures — superlattices.[16,17] The frequencies of folded acoustic phonons in a periodic layered structure are determined only by the average period of a system and are independent on the internal structure of layers. Hence the positions of doublets are the same for planar superlattices and structures with three-dimensional islands having the same period. The in-plane irregularity caused by island formation only leads to the decrease of doublet intensity.

As one can see in Fig. 2, the second and third doublets of folded LA phonons centered at 31.5 and 47 cm^{-1} are clearly observed in the spectra of AlAs/InAs structure (sample *D*). The first doublet should appear close to the laser line at about 16 cm^{-1}, but it is not seen because of the strong background. Although our samples contain only five periods, it is enough for folded LA doublets to appear. For GaAs/InAs structures the doublets are not so clear probably due to the smaller amplitude of acoustic and photoelastic

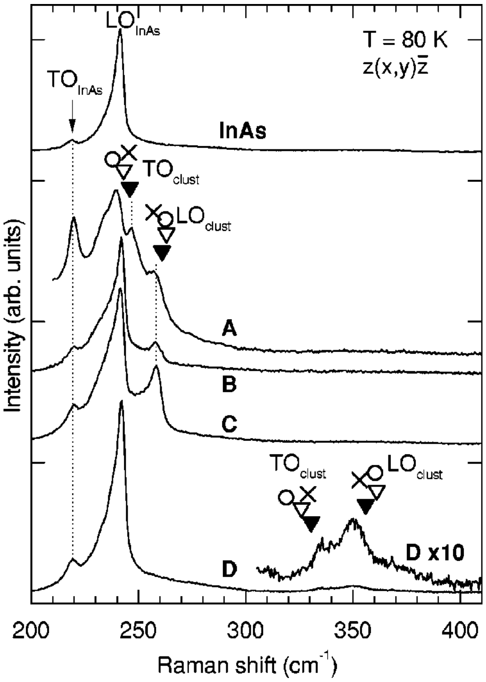

FIG. 3. Raman spectra of samples *A*–*D* in the region of optical phonons. Symbols show calculated phonon frequencies in coherently strained GaAs and AlAs clusters having the shape of spheres (circles), pyramids (open triangles), truncated pyramids (solid triangles), and pseudomorphic GaAs and AlAs layers (crosses). Dashed lines are guides to the eye.

modulation in the GaAs/InAs system compared to that for AlAs/InAs. The Raman intensity of folded acoustic phonons in a periodic layered structure is proportional to the difference of photoelastic coefficients of the constituents.[16]

Triangles in Fig. 2 indicate the positions of folded LA lines calculated using the elastic continuum model[16,17] for GaAs/InAs and AlAs/InAs multilayer structures with layer thicknesses equal to nominal ones for the samples *A* and *D*. As one can see in Fig. 2, the calculated frequencies agree well with experimentally observed peaks of folded LA doublets.

The features at 90, 106, 154, and 165 cm^{-1} are due to the second-order Raman scattering by acoustic phonons of bulk InAs.[18] The broadband of disorder-activated InAs LA phonons appears at about 140 cm^{-1}. Assignment of these features according to Ref. 18 is given in Fig. 2. They were also observed in the spectra of the bare InAs substrate.

B. Optical-phonon region

Raman spectra of samples *A*–*D* in the region of optical phonons are shown in Fig. 3. The spectrum of a bare InAs substrate is also given as a reference. The peaks of InAs longitudinal-optical (LO) and transverse-optical (TO) phonons at 242 and 219 cm^{-1} are present in all spectra. The asymmetric line shape of the InAs LO-phonon peak is due to the influence of free carriers in this narrow-gap material.[19] In sample *A*, the InAs LO-phonon line appears slightly downward-shifted and decreased in intensity, while the TO peak intensity grows. This is probably due to the increased degree of structural disorder or possible alloying in this sample having the largest amount of deposited GaAs. It should be noted that because of the strong absorption of InAs at the wavelength used in our experiments (514.5 nm), the substrate contribution to the Raman spectra is negligible. Practically all the signal due to InAs phonons originates from the InAs spacer layers between GaAs (AlAs) clusters. The appearance of an intensive forbidden TO line in this sample also indicates the presence of disorder. Possible alloying can cause a slight shift of the InAs TO-phonon peak towards higher frequencies[20] observed in sample *A* (about 1 cm^{-1}, which can hardly be seen in Fig. 3).

Besides the InAs phonon peaks, the spectra of samples *A*–*C* contain the LO-phonon line of GaAs clusters. The frequency position of this line is practically the same for samples *B* and *C* (259 cm^{-1}), independent on the amount of deposited GaAs. In sample *A* the LO-phonon line of GaAs clusters is again slightly shifted, and the TO-phonon line of GaAs clusters is also seen at 247 cm^{-1}. No peaks were observed at the positions of bulk GaAs LO and TO phonons (295 and 271 cm^{-1}, respectively, for $T=80$ K). Similar behavior (significant downward shift of cluster phonon lines related to bulk positions) was observed for the spectra of the sample with AlAs clusters (*D*). The LO- and TO-phonon lines of AlAs clusters appear at 350 and 335 cm^{-1} while the bulk positions are 405 and 363 cm^{-1}, respectively.

Two main factors can cause the phonon frequency shift in perfect nanostructures: strain and confinement. In our case the influence of confinement on the cluster phonon frequencies is small. For such large GaAs clusters, the phonon frequency shift due to confinement in the growth direction estimated from the bulk GaAs optical-phonon dispersion[21,22] is about 1–2 cm^{-1}. Since the lateral dimensions of clusters are much larger than their height, lateral confinement effects can be neglected. In the case of AlAs clusters, the frequency shift induced by confinement is even smaller because of weak dispersion of AlAs optical phonons.[21,23] The difference between the position of the LO-phonon line in the spectra of AlAs clusters and the corresponding bulk frequency in unstrained AlAs is 55 cm^{-1}. This is about six times larger than the whole dispersion of AlAs LO phonons in the (001) direction[21] (8–10 cm^{-1}). Therefore, such a large phonon frequency shift as observed in our spectra can only be explained by the presence of mechanical strain in AlAs (GaAs) clusters.

The strain-induced shifts of optical-phonon frequencies in diamond- and zinc-blende-type crystals are determined by the solutions of the secular equation given by Cerdeira *et al.*[24] In particular, for the nanostructures grown on (001)-oriented substrates, the strain dependence of phonon frequencies is given by the following equations:

$$p\varepsilon_{zz}+q(\varepsilon_{xx}+\varepsilon_{yy})=\omega^2-\omega_0^2 \tag{1}$$

for LO phonons and

$$p\varepsilon_{xx}+q(\varepsilon_{yy}+\varepsilon_{zz})=\omega^2-\omega_0^2 \tag{2}$$

for TO phonons. Here ε_{xx}, ε_{yy}, ε_{zz} are the diagonal components of the strain tensor, and ω and ω_0 are the phonon frequencies in strained and unstrained material, respectively. The shear strain components ε_{ij} $(i\neq j)$ are neglected due to the reason outlined below. *p* and *q* are the phonon deformation potentials that determine the strain dependence of phonon frequencies. The values of p and q for GaAs were taken from Ref. 25. For AlAs, to the best of our knowledge, there are no data about the phonon deformation potentials. To estimate the phonon frequencies in strained AlAs clusters, we assumed the values p/ω_0^2 and q/ω_0^2 to be equal to those in GaAs. Such an approximation is reasonable because these parameters can be expressed in terms of effective interatomic force constants.[24] According to *ab initio* calculations of phonon dispersions,[21] the force constants of GaAs and AlAs are very similar, and the bulk AlAs phonon spectrum is well described by a calculation using GaAs force constants. The difference in phonon frequencies of these two materials is mainly caused by large difference of cation masses (mass approximation). Elastic constants of GaAs and AlAs are also almost equal [the difference is less than 5% (Ref. 26].

Strain fields in QD's have been calculated by several groups.[5,12,27–29] Grundmann *et al.*[5] have calculated the strain distribution in InAs QD's embedded in GaAs and estimated the phonon energies in these QD's. Since optical phonons are confined within the whole cluster, their frequencies are determined by average components of the strain tensor. The shear strain has been shown to be significant near the edges of QD's and small inside the islands.[12,29] Moreover, the average values of shear strain components vanish due to the symmetry requirements.[12] [We assume our clusters to have biaxial symmetry in the (x,y) plane.] Rewriting Eqs. (1) and (2) in terms of an average hydrostatic strain component $\varepsilon_h = \varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}$, and a biaxial strain component $\varepsilon_b = \varepsilon_{zz} - (\varepsilon_{xx} + \varepsilon_{yy})/2$, we have calculated the phonon frequencies in GaAs and AlAs clusters embedded in InAs. We used average values ε_h and ε_b obtained from strain distributions for InAs quantum dots of different shapes embedded in GaAs given by Andreev *et al.*[29] In order to test the way we perform our calculations, we have obtained the LO-phonon energy for pyramidal InAs islands in the GaAs matrix. The calculated energy is 32.5 meV, which agrees well with the value 32.1 meV obtained by Grundmann *et al.*[5] for these QD's by averaging the optical-phonon energies calculated at each point of QD for respective local strain. The experimental value determined by resonant photoluminescence[2,4] is 32.2 meV.

To obtain correct values of ε_h and ε_b in GaAs (AlAs) clusters embedded in InAs, the opposite sign of misfit strain was taken into account [$\varepsilon_0 = 0.0716$ instead of -0.067; $\varepsilon_0 = (a_M - a_{\mathrm{cl}})/a_{\mathrm{cl}}$, where a_M and a_{cl} are lattice constants of matrix and cluster materials, respectively).

Calculated optical-phonon frequencies for GaAs and AlAs clusters having the shape of sphere, pyramid, and truncated pyramid are shown in Fig. 3 by symbols. We have also calculated the strain-induced shifts of phonon frequencies for pseudomorphic GaAs and AlAs layers embedded in InAs (confinement effects were left out of account here) taking $\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon = 0.07$, $\varepsilon_{zz} = 2\varepsilon S_{12}/(S_{11} + S_{12})$, where S_{11} and S_{12} are elastic compliance constants taken from Refs. 24 and 26. As one can see, the results of the calculation correspond well to the experimental positions of phonon lines in the spectra. This evidences that clusters are coherently strained, i.e., there are no dislocations leading to strain relaxation. This fact is also confirmed by atomic resolution TEM images. The best agreement between experimental and calculated phonon frequencies is obtained for clusters having the shape of a truncated pyramid. From TEM data (Fig. 1) one can see that islands indeed seem to have such a shape. Thus, Raman spectra can be sensitive to the shape of self-assembled islands, as it determines the average strain components. However, more sophisticated calculations of strain tensors and phonon frequency shifts (due to both strain and confinement) are required to make more reliable conclusions from Raman spectra.

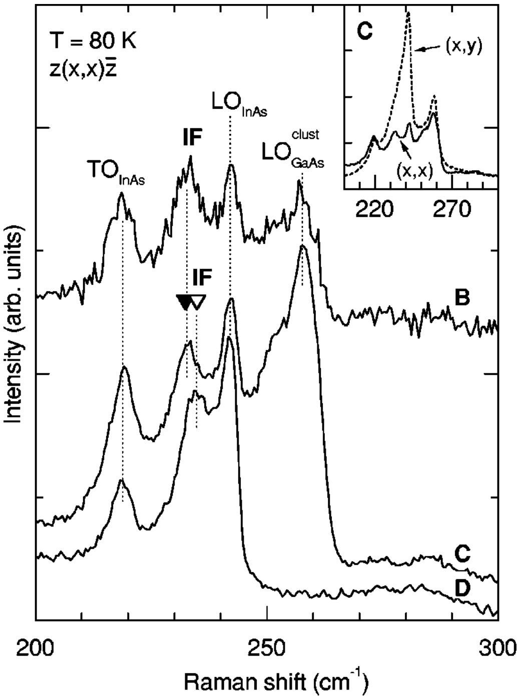

FIG. 4. Optical-phonon Raman spectra of samples $B,C,$ and D in parallel polarization configuration. Solid and open triangles show the calculated frequencies of the first interface modes for GaAs and AlAs clusters, respectively. Dashed lines are guides to the eye. Inset shows the relative intensities of the spectra in parallel (x,x) and crossed (x,y) polarization geometries for sample C.

We have not observed any features that can be attributed to the vibrations of a wetting layer (WL). In the case of GaAs clusters, this is possibly due to the confinement effect, which is large in a thin wetting layer. The phonon lines in a GaAs WL should be moved towards lower frequencies by about 10 cm^{-1} in addition to the strong strain-induced shift. Hence, these lines can be close to the strong InAs LO-phonon peak and are therefore not distinguishable in the Raman spectra. In the case of AlAs clusters, the WL phonon lines were not observed probably due to their small intensity. TEM images (Fig. 1) do not give unambiguous evidence of the presence (or not) of a wetting layer. However, they show that even for small amounts of deposited GaAs or AlAs (samples C and D), most of it is contained in clusters. It should be noted that according to recent calculations,[12] the presence of WL does not significantly influence the strain field inside the islands.

Figure 4 shows the Raman spectra of samples $B,C,$ and D in the region of InAs optical phonons measured with parallel polarizations of incident and scattered light. The intensity of the InAs LO-phonon peak, as expected, strongly decreases compared to the spectra measured in crossed polarization geometry shown in Fig. 3, while the LO-phonon line of the

GaAs clusters is only slightly weaker. (See the inset of Fig. 4, where the spectra of the sample *C* measured in two configurations are given for comparison.) This indicates that the role of scattering via the Fröhlich mechanism of electron-phonon interaction which causes the polarized Raman scattering by LO phonons[16,30] is more significant in nanoclusters compared to bulk material.

A new feature appears between lines of InAs TO and LO phonons in the parallel polarization gepmetry. For all the samples with GaAs clusters (A–C), the position of this feature ($233\ \mathrm{cm}^{-1}$) is independent of the amount of deposited GaAs. In sample *D* containing AlAs clusters, this line is shifted to $235\ \mathrm{cm}^{-1}$. We attribute this feature to InAs-like interface phonons. These are vibrations localized near the interfaces. Such modes exist in the frequency ranges of both cluster and matrix material. Using the dielectric continuum model,[31] we have calculated the frequencies of the first InAs-like interface phonons for spherical GaAs and AlAs clusters embedded in InAs. The results of this calculation correspond well to the experimental data and predict correctly the frequency shift of InAs-like interface phonons in a sample with AlAs clusters relative to those in GaAs/InAs structures. This shift is determined by the difference in frequency-dependent dielectric functions of GaAs and AlAs.

It should be noted that in a planar layered structure (superlattice) with a high ratio of layer thicknesses, as in our samples (nominally 0.6–1-nm-thick GaAs or AlAs layers separated by 10-nm InAs spacers), interface phonon bands appear in narrow intervals at $1–2\ \mathrm{cm}^{-1}$ from bulk TO and LO phonons.[16,32] The possible presence of wetting layers having planar interfaces with the matrix material can also lead to the appearance of interface modes close to bulk InAs LO and TO phonons. Therefore, the presence of interface-phonon features approximately in the middle between InAs TO and LO phonons is another consequence of three-dimensional island formation.

Comparison of calculations with experimental data shows that there is no significant influence of strain on the observed InAs-like interface phonons. Pusep *et al.*[6] observed higher-order GaAs-like interface phonons in resonant Raman spectra of InAs QD's embedded in GaAs and found that these modes can be influenced by strain. The higher-order interface modes are localized near apexes of pyramidal InAs QD's where strain has sharp maxima. Our clusters are unlikely to have such sharp tips, and we observed only the first InAs-like interface mode. It is not strongly localized near the interface and propagates on both sides of it, where the strain has opposite signs. Therefore, this mode appears to be less strain sensitive compared to higher-order interface phonons.

IV. CONCLUSION

The phonon spectra of GaAs and AlAs self-assembled nanoclusters in an InAs matrix were investigated by Raman spectroscopy. Doublets of folded acoustic phonons are present in the low-frequency Raman spectra of multilayer structures with GaAs and AlAs islands. The frequencies of these doublet peaks agree well with elastic continuum model calculations and correspond to the folded acoustic phonons of superlattices with the same period. This shows that three-dimensional island formation does not affect significantly the low-frequency acoustic phonons.

In the optical-phonon region we observed large strain-induced shifts of phonon lines of GaAs and AlAs clusters from respective bulk values. We have calculated the phonon frequencies in strained clusters of different shape. According to the calculations, the experimentally measured optical-phonon frequencies are characteristic for coherently strained dislocation-free GaAs and AlAs islands. Experimental data are described in the best way by calculations performed for clusters with the shape of a truncated pyramid. The absence of dislocations in the samples studied is confirmed by transmission electron microscopy.

The features of InAs-like interface phonons were observed in polarized Raman spectra. The frequency positions of these features correspond well to the calculations based on the dielectric continuum model and also give evidence of three-dimensional island formation.

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*Electronic address: tenne@thermo.isp.nsc.ru

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