Lattice dynamics of superlattices with interface roughness

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We present and analyze a lattice-dynamics calculation of superlattices taking into account interface imperfections. We get a quantitative description of the change in the confinement shift due to a small-scale interface roughness. We also introduce a critical size in the lateral scale of this roughness above which Raman-line splittings should be observed.

A great amount of work has been devoted in the past few years to the growth and study of very-thin-layer superlattices based on the GaAs/AlAs,[1] GaAs/InAs,[2] and Ge/Si (Ref. 3) couples of bulk constituent materials. It is a very difficult task to get an estimation of the degree of intermixing around each interface which arises from the growth statistics. It has to be reduced drastically when attempting to grow individual layers as thin as 1 or 2 monolayers. Moreover, it is of great interest to determine the spatial characteristics of the roughness: its amplitude along the growth axis and the dimension of the involved fluctuations parallel to the layer plane. Experimental information on these parameters has already been obtained from transmission electron microscopy (TEM),[4,5] high-resolution x-ray diffraction[6] (which is well adapted to the observation of short-range fluctuations) and luminescence on excitonic recombinations,[7,8] which probes long-range fluctuations, often called terraces. We show in this paper, on the basis of a three-dimensional (3D) lattice-dynamics model including the interface roughness, that confined optical vibrations are very sensitive to both amplitude and lateral distribution of the roughness. As the different confined eigenmodes probe different lateral extensions, Raman scattering should become a unique tool to further investigate the size of the interface terraces and to characterize the novel lateral superlattices.[9]

It is now well established[10] that the optical vibrations in these structures are strongly confined either in the GaAs or AlAs layers and that their frequencies reflect the boundary conditions at each interface. Several lines actually appear in the Raman spectra, associated to successive longitudinal-optical (LO) vibrations confined in the GaAs layers. The corresponding modes for the AlAs layers are much less Raman active and will not be considered in what follows. The confined frequencies can be obtained, at least in perfect samples with abrupt interfaces, from the only knowledge of the individual GaAs-layer thickness. They correspond to the bulk LO frequencies at a few well-defined finite wave vectors:

$$k_s^1 = \frac{s\pi}{(n_1+1)a} , \tag{1}$$

where a is the monolayer thickness and n_1 the number of monolayers in the GaAs layer.

When interface broadening is present, one observes[11] an increase in the confinement shift of the LO phonons which can be associated, on the basis of the previous analysis, with a decrease of the effective layer thickness "seen" by the confined vibrations. This result qualitatively reflects the formation of an intermediate alloy layer which pushes away the vibrations closer to the center of the layer in which they are confined. We previously obtained a good quantitative description of this effect using a one-dimensional (1D) lattice-dynamics model with the average Al contents varying from plane to plane around the interface. The use of a 1D model is justified when shifts of the confined LO Raman lines are observed due to interface roughness, but insufficient when splittings appear.[12] The corresponding modes then become sensitive to the presence of terraces of either pure GaAs or pure AlAs in a way similar to the case of excitonic recombination lines.[7,8] In this paper we investigate, on the basis of a three-dimensional lattice-dynamics model including interface roughness, the respective conditions for observing either a gradual interface regime, where the conclusions of 1D models are valid, or a terraced interface regime. This will lead us to reexamine the few presently available relevant Raman-scattering results.

3D lattice dynamics is easy to compute for GaAs/AlAs structures because of the negligible difference in the force-constant matrices of both constituents. We use in this work the matrix determined in Ref. 13 for GaAs in the frame of the overlap-valence-shell model. The force-constant matrix on the supercell at a given wave vector in the Brillouin minizone is then deduced[14] from the basic unit-cell ones at the wave vectors in the full Brillouin zone which are now equivalent due to the modulation. To introduce interface broadening, we multiply the unit cell in two different cubic directions: z, which is the superlattice axis, and x, which is in the plane of the layers. When assuming flat interfaces, the Ga and Al atoms are distributed periodically along the z direction and a given layer perpendicular to z contains only one atomic species. To model interface roughness, we also modulate the site occupation along the x direction in one or more layers near the nominal interface plane separating GaAs from the previously grown AlAs. The

resulting interface looks like corrugated cardboard. The other interface (AlAs on GaAs) is assumed to be abrupt on the basis of several experimental analyses.[8] The corresponding unit cell, illustrated in Fig. 1(a), allows us to incorporate roughness in the lattice dynamics while remaining within the present computational limitations.

Using this model we are able to calculate the lattice dynamics of superlattices with noninteger individual layer thicknesses, $(GaAs)_{n_1+(1-x)}/(AlAs)_{n_2+x}$. We investigate the effect of different interfacial statistics by varying the in-plane period $p+q$ between 2 and 40 monolayer thickness, i.e., between 6 and 120 Å. p and q are the respective numbers of adjacent Ga and Al atoms in the single interfacial layer [see Fig. 1(a)]. The average aluminum content x in the interface layer then takes simple rational values $q/(p+q)$, ranging from 0 to 1. While a single composition $x=0.5$ can be considered for $p+q=2$, a series of nine different ones is considered for $p+q=10$. The frequency of the higher-energy ($s=1$) confined mode in the GaAs layers, calculated at a vanishingly small wave vector along the superlattice axis, is shown on Fig. 1(b) for $n_1=n_2=2$ and for several choices of x and $p+q$. The results display very similar trends for other nominal thicknesses, except that the absolute energy variations decrease with increasing n_1.

Let us first consider the points on the lowest curve of Fig. 1(b). They correspond to Al concentrations 0.1, 0.25, 0.5, 0.75, and 0.9, and the in-plane atomic distribution in the interface layer is such that isolated Ga or Al atoms are separated by a distance comparable to that in a 1D random alloy of the same composition. These points thus give us an estimation of the effect of an intermixed layer with short-range (alloylike) disorder. Between $x=0.5$ and 1, the frequency of the confined vibrations remains remarkably close to that obtained with exactly two monolayers. Even only one Al atom on each fourth site ($x=0.25$) almost completely pushes away the vibration out of the interfacial layer. This result is in quite good agreement with the predictions of the 1D model.[11]

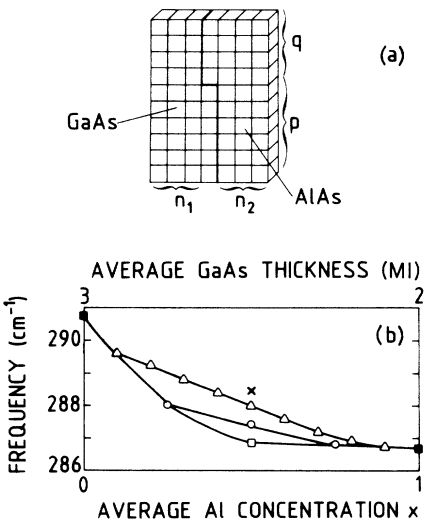

FIG. 1. Frequency [panel (b)] of the fundamental GaAs-type LO vibration [$s=1$ in Eq. (1)] calculated for a $(GaAs)_{2+(1-x)}/(AlAs)_{2+x}$ superlattice with the supercell schematized in (a), as a function of the average aluminum concentration x in the in-plane modulated interface atomic layer. Different periods of the in-plane modulation are considered: $p+q=2$ (open square), 4 (open circles), 10 (open triangles), and 40 (cross). The corresponding average thickness $2+(1-x)$ of the GaAs layers is indicated on the upper scale. The solid lines are only a guide to the eye.

We can also note in Fig. 1 the variation of the $s=1$ frequencies as a function of the size of the terraces for a given average concentration. Let us consider the case where $x=0.5$ and vary $p=q$ from 1 to 20. The frequency then slightly shifts towards higher frequency while the eigendisplacement is weakly modified. No line splitting is predicted up to this high value of the terrace size. This result is consistent with the absence of any reported observation of such splitting. It is, however, surprising by comparison with the behavior of electrons confined in a GaAs layer with rough interfaces.[15] Well-defined electronic levels, associated with the lowest quantized levels in each thickness, were indeed predicted to appear when the terrace extension exceeds the following dimension:

$$\lambda = d\sqrt{m_z/m_{xy}}/s\sqrt{2\,\Delta d/d}\ , \tag{2}$$

where m_z and m_{xy} are the effective masses along z in the bulk constituent (which governs the confinement in quantum wells with infinite barriers) and along the layer plane in the superlattice; d and Δd are the nominal thickness and its fluctuation, and s is the index of the quantized level. This expression originates in the comparison between the additional confinement energies due to either the reduction of the layer thickness or the lateral localization in the thicker parts. Assuming an isotropic mass, i.e., starting from a cubic crystal and assuming that the confinement does not significantly modify m_{xy}, this length scale does not depend on the actual value of the mass, but only on the geometry of the problem. It increases with increasing nominal layer thickness and with decreasing defect amplitude. A naive application of this criterion to the confined vibration which we analyzed previously would let us predict the emergence of lateral localization for very small terraces (3 and 9 monolayers for $n_1=2$ and 5, respectively), in complete disagreement with the predictions of our calculation.

We attribute this disagreement to the long-range Coulomb forces which strongly affect the dispersive properties of optical phonons around the zone center.[16] We show in Fig. 2 the dispersion of the GaAs-type optical phonons with displacement mainly oriented along z, calculated at a fixed finite wave vector along z, $k_z=0.01$ in reduced units, and an in-plane wave vector k_x varying between 0 and 0.3. The dispersion curve of the fundamental mode is rapidly varying close to the zone center due to the increasing associated macroscopic polarization. As a result of this huge anisotropy, the lateral localization does not appear up to very large terrace size. On the contrary, this macroscopic polarization remains very small for the other odd confined modes and is vanishing for the even ones. As a consequence, their in-plane dispersion curves are smooth close to the zone center. We thus pre-

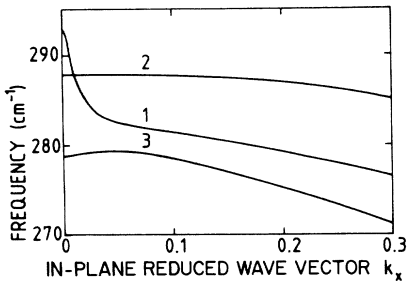

FIG. 2. Dispersion curves in the layer plane of the higher-frequency GaAs-type optical vibrations calculated for a $(GaAs)_5/(AlAs)_3$ superlattice with perfect interfaces.

dict that the corresponding confined modes should be much more sensitive to interface roughness.

We show in Fig. 3 some eigendisplacements in the supercell calculated for a corrugated structure with $n_1=5$ and $n_2=3$ and corresponding to the higher even mode [$s=2$ in Eq. (1)]. We have clear evidence in this case of a

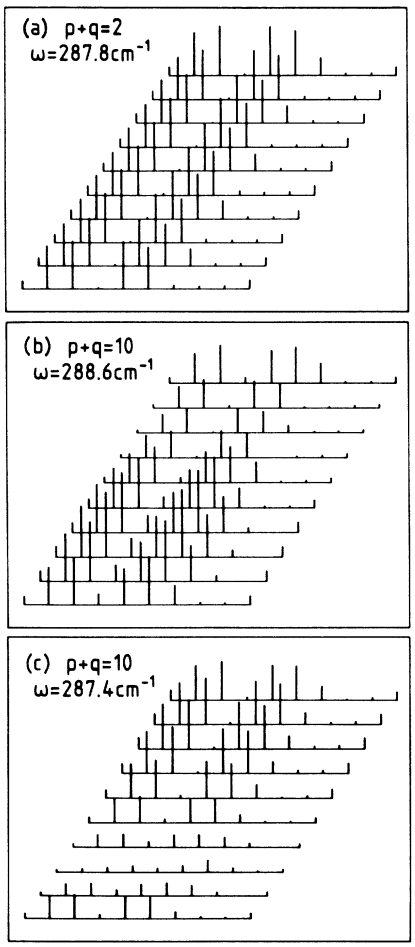

FIG. 3. Amplitude of the eigendisplacements along z corresponding to the second GaAs-type LO vibrations [$s=2$ in Eq. (1)], shown on the cation sites in the supercell of a $(GaAs)_{5.5}/(AlAs)_{3.5}$ superlattice with two different in-plane modulations: (a) $p+q=2$; (b) and (c) $p+q=10$. In the latter case two different eigenmodes are displayed, as explained in the text.

mode splitting due to lateral terraces, when they reach a large enough extension. In Fig. 3(a), we show the eigendisplacement obtained assuming a small in-plane period of 2 monolayers. There is almost no modulation along the x direction, the displacement remaining hardly distinguishable from that in the perfect $n_1=5$ GaAs layer. In particular, the displacement of the Ga atom at the center of this perfect well is vanishing by symmetry and remains negligible in the locally 6-monolayer-thick parts of the rough well. In good agreement with a critical size $\lambda=4$ deduced from relation (2), opposite conclusion applies to the sample with larger terraces: two different eigenmodes issue from the $s=2$ mode of the perfect GaAs layer and exhibit a single node in the vicinity of the center of the layer. The lowest-frequency component of the doublet is partially localized in the narrow parts of the GaAs layer, and the highest one, more clearly, in the wide parts. A good signature of this difference, again, is obtained from the displacement of the Ga atom at the center of the narrow parts. Its displacement is vanishingly small for the lowest mode, but becomes significant in the wide parts for the highest-frequency vibration. The node indeed is now shifted from the gallium site to the neighboring arsenic one.

Similar behavior can be evidenced for the highest-index ($s>2$) confined vibrations, with the qualitative tendency to a decrease of the critical terrace size with increasing s value. On the other hand, this critical size for a given value of s increases with increasing nominal thickness n_1. For instance, mode 2, which is split in the sample of Figs. 3(b) and 3(c), becomes delocalized when n_1 is increased from 5 ($\lambda=4$) to 10 ($\lambda=12$), the terrace size remaining unchanged. All these variations reflect the change in the in-plane perturbative potential, and the value of λ thus appears to be an excellent criterion of the vibrational behavior. The analysis should therefore remain valid in the more realistic case of two-dimensional (2D) aperiodic fluctuations. Since the optical-phonon dispersion is isotropic in the layer plane, one should predict a moderate increase of the critical parameter when replacing 1D fluctuations by 2D ones (by a factor $\sqrt{2}$ for square defects). A typical fluctuation scale of 10 monolayers should allow the observation of splittings of modes 3 and 5 in 10-monolayer-thick GaAs layers. Our previous experimental results[11] and more recent ones with growth temperatures as low as 400 °C (Ref. 17) strongly suggest the persistence, even in excellent samples, of a very-small-scale roughness whose distribution along the growth axis is closely related to the growth temperature. This conclusion is in good agreement with some TEM observations[4,5] and stresses the importance of atomic segregation to limit the abruptness of the heterointerfaces.[18] Raman scattering on confined vibrations should become a powerful tool in assessing growth conditions which circumvent this thermodynamic limitation.

In summary, we presented in this paper the predictions of a lattice-dynamics calculation of superlattices with interface imperfections. We modeled these imperfections through a one-dimensional periodic corrugation in the interface plane. Despite this crude approximation, our calculation brings about interesting insight into the effect of

interface roughness on the confined phonon frequencies. Moreover, it should be directly useful for analyzing the Raman spectra on the recently introduced[9] lateral superlattices. We saw evidence of the transition between two different behaviors when the lateral size of the fluctuations is increased. For very small terraces, a single mode is extended over the whole layer plane is and mostly confined in the central part of the layer, which is less affected by the roughness. For larger terraces, this single mode splits into two different ones, localized along the layer planes in regions of different local thickness. This critical size depends on the average thickness of the layers, on the depth of the fluctuations, and on the considered vibration. It is generally small, in the 10-Å range, with the remarkable exception of the fundamental vibration, which remains extended up to long in-plane modulations. Raman-scattering investigations of these fluctuations should therefore be focused on higher-index modes in moderately thin individual layers. Let us finally stress that the criterion introduced in this paper for confined vibrations should be applicable to other quasiparticles confined in imperfect quantum wells.

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