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Interfaces, confinement, and resonant Raman scattering in Ge/Si quantum wells

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We address the question of confinement of the Ge-Ge mode in 5-ML-Ge single and multiple quantum wells. Using Raman scattering, our data show strong dependence of the interface quality on the number of quantum wells and thereby on the confinement of both the phonons and the electronic states in the Ge wells. The dependence of line shape and peak position of the Ge-Ge Raman line with laser photon energy gives a clear indication of the existence of terraces in the interfaces of the Ge/Si multiple quantum wells.

I. INTRODUCTION

The usefulness of studying the phonon spectrum in order to probe the interface structure of GaAs/AlAs superlattices has been demonstrated both experimentally and theoretically.[1–4] Early Raman scattering studies in Ge_nSi_m superlattices, together with simple supercell calculations, suggested that the Ge-Si vibration in these structures could be used as an indication of interfacial quality.[5] More recently, this suggestion has been extensively studied by both theoretical and experimental methods.[6–9] These studies indicate that the small-scale roughness can be simulated by placing a few monolayers of Ge_xSi_{1-x} alloy at the Ge/Si interface. In bad interfaces, a thick alloy layer of graded composition would separate the regions of pure Ge and pure Si. In "good" but realistic interfaces, 2 ML (one on each side of the ideal Ge/Si interface) of composition $x \sim 0.5$ should give a reasonable picture of the small-scale roughness of the Ge/Si interface. This interpretation is thoroughly explored in Refs. 6 and 7, and successfully used in Ref. 9 to interpret Raman scattering data from a variety of Ge_nSi_m strain-symmetrized superlattices. There is another type of interface imperfection that is found in III-V superlattices interfaces, namely the existence of terraces, several hundred angstroms in linear dimensions, of different widths, which lead to a splitting of the exciton line in the photoluminescence spectrum.[1,10] Although such direct evidence of terracing would be hard to find in Ge/Si microstructures (owing to their indirect gap), this type of large-scale roughness could manifest itself in the Raman spectrum through line-shape variations in the phonon lines, as a recent theoretical work by de Gironcoli and Molinari[8] suggests. In the present work, we study the Raman spectrum of samples composed of one, two, and six Ge_5 quantum wells separated by 5-ML-thick Si barriers (in multiple quantum wells) and bounded by thick (300 Å) Si spacer layers. By studying the dependence of the position and shape of the Raman peaks originating in Ge-Ge and Ge-Si vibrations in the different samples for a fixed laser frequency and in a given sample for several laser frequencies, we obtain evidence of the coexistence of both types of interface roughness (small-scale, alloying, and large-scale, terracing) in the Ge/Si interface.

II. EXPERIMENTAL DETAILS

The samples were grown by molecular beam epitaxy at low temperatures on Si(100) substrates.[11] They consist of a single (1QW), a double (2QW), and a sextuple (6QW) quantum well made of five Ge monolayers separated (in multiple wells) by Si barriers of 5 ML. Each structure is repeated 20 times (1QW and 2QW) and 10 times (6QW) and each unit is separated from the other by 300-Å Si spacer layers. The structures are grown on a Si buffer layer and their in-plane lattice parameter is that of bulk Si, so the Ge quantum wells are under biaxial compression.

Room temperature Raman data were taken using a Dilor Triplemate micro-Raman System with a charge-coupled-device detector. Exciting radiation was obtained from the lines of an Argon-ion laser and the 633-nm line of a He-Ne laser focused by the microscope objective (×100 magnification) at low-power densities in order to prevent sample heating and free carrier excitation. When required, a reducing confocal opening was used in order to diminish the sample area probed. All spectra were taken in the backscattering configuration with light incident either along or perpendicular to the growth axis. In the latter case a cleavage face was used. The configurations used were $z(x,y)\bar{z}$, $y'(x',x')\bar{y'}$, and $y'(x',z)\bar{y'}$,

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where the x, y, z axes are the cubic crystallographic axes with $z = [001]$ as the growth direction, $y' = [110]$, and $x' = [\bar{1}10]$ are axes perpendicular and parallel to the cleavage face, respectively. In the first two configurations, the Raman peaks originate in vibrations along z while in the $y'(x', z)\bar{y}'$ configuration only in-plane vibrations are allowed. In accordance with previous usage,[6–9] we shall call the first longitudinal optical (LO) vibrations and the second transversal optical (TO) modes, regardless of their longitudinal (L) or transverse (T) character in relation to the direction of propagation. This is because in quantum wells and superlattices vibrations along (perpendicular to) the quantization (growth) axis are related to the bulk LO-phonon (TO-phonon) dispersion independent of the propagation direction.[12] In accordance with this, the spectra obtained in the $z(x, y)\bar{z}$ and the $y'(x', x')\bar{y}'$ configurations are identical for a given sample and laser excitation frequency.

III. RESULTS AND DISCUSSION

The Raman spectra for the LO vibrations of the samples used in this work was first reported by Rodrigues *et al.*[13] Three main optical lines are observed, which are known as the Ge-Ge mode ($\sim$ 310 cm^{-1}), the Si-Si mode ($\sim$ 510 cm^{-1}), and the Ge-Si mode ($\sim$ 415 cm^{-1}). Figure 1 shows spectra of the 6QW sample, where these three structures are seen together with the bulk Si line ($\sim$ 520 cm^{-1}) from the buffer and spacer layers. The analogies and differences between these modes and similarly labeled optical modes in bulk Ge_xSi_{1-x} alloys[14] are explicitly shown in Fig. 4 of Ref. 13. The alloy modes have been recently discussed in detail in the light of several types of model calculations.[15,16] In particular, de Gironcoli and Baroni[16] show that their designation corresponds to the type of bond which contributes to each type of vibration. These authors emphasize the importance of local environment in determining the position and shape of the lines appearing in the Raman spectrum. This is particularly true for the Ge-Si LO line, where the line is broad, asymmetric, and has several satellite structures towards its high-energy end. This is believed to be a consequence of the contributions of different Ge-Si clusters.[15,16]

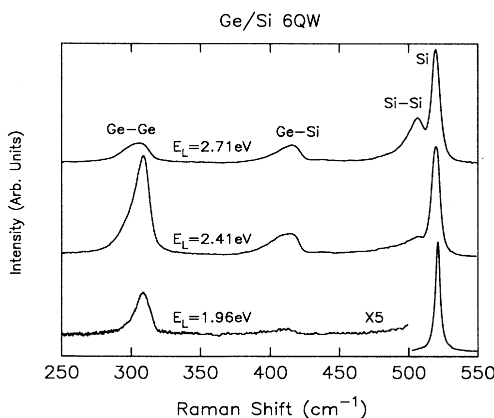

FIG. 1. The Raman spectra, in the LO phonon region, of the 6QW sample for different laser photon energies.

In a superlattice with perfectly sharp interfaces, a narrow line should appear at a frequency that is lower than that of the alloy Ge-Si mode, but higher than that of the Ge-Ge vibration, for configurations in which only LO modes are allowed.[5] Although the main contribution to this LO-like vibration comes from vibrations of Si against Ge atoms across the interface, there is also some contribution from Ge vibration away from the border, so it can only be approximately called an interface mode.[5] On the other hand, for the TO polarization, true interface modes are allowed.[6–9] They would also result in narrow lines at lower frequencies than those actually observed in the Ge-Si vibrations of either alloys or superlattices.[5,9,13,14] Instead, the observed Ge-Si LO lines in Ge/Si quantum wells and superlattices resemble in every respect those of Ge_xSi_{1-x} alloys.[5–7,9] For the T polarization, however, the observed line shapes are more complex and seem to contain mixtures of alloylike and ideal-interface peaks.[6,7] These features of the Ge-Si vibrations in Ge_nSi_m superlattices leads to associating the small-scale roughness of the interface to the existence of a few monolayers of Ge_xSi_{1-x} alloy, separating the pure Si from the pure Ge regions. Alloying at the interface was also invoked for the same purpose in GaAs/AlAs superlattices.[1–4] To the best of our knowledge, the second type of interface roughness found in those cases,[1,10] namely, terracing on the several hundred angstroms scale, has not been experimentally observed in Ge/Si quantum wells and superlattices. We propose to call attention to this aspect of interface roughness and its effect on the Raman spectrum of these materials. de Gironcoli and Molinari[8] calculate the effect of terracing on the Si-Si LO vibration of strain-symmetrized Ge_nSi_m superlattices. Their calculation includes a periodic arrangement of terraces, which leads to a splitting of this Raman line for terraces of linear dimensions

$$D \geq d(d/2\Delta d)^{1/2}, \tag{1}$$

where d and Δd are the layer thickness and its fluctuation, respectively. In our case, this would mean a lateral size $D \geq 8$ ML for a fluctuation of 1 ML. In a real interface, a distribution of thickness (Δd's) would be encountered and a broadened line, possibly asymmetric, would be observed. For this reason, the test for terracing proposed by these authors may not be useful in practice. On the other hand, if the terraces were of linear dimensions comparable to or larger than the excitonic radius,[1,10] resonant Raman scattering could be used to probe selectively each terrace. For this to happen, the vibration studied and the exciton in resonance with the laser frequency should be confined within the same terrace. Changing the laser line would bring different terraces into resonant conditions and thus enhance the contribution of vibrations at different frequencies. The global effect would be a marked variation of line shape as the laser frequency is varied around such a resonance. In what follows, we pursue this idea systematically. Instead

of the Si-Si line we use the Ge-Ge line because a resonance such as the one described above exists in an accessible region of the optical spectrum ($E_L \sim 2.5$ eV). The Si-Si line is more difficult to use because its resonance occurs towards the UV part of the spectrum,[13,17,18] and also because the proximity of the much stronger bulk Si line makes line-shape studies difficult.

In Fig. 1 we show spectra, in the optical phonon region, of the 6QW sample taken in the $z(x,y)\bar{z}$ configuration for three different values of incident laser frequency. The effect of changing the excitation energy on the intensity of the different peaks (resonant Raman scattering) of the Raman spectrum of the samples used in the present work is discussed qualitatively in Ref. 13 and with more detail in Ref. 17, while resonant Raman scattering in Ge_nSi_m strain-symmetrized superlattices is reported in Ref. 18. The effects described in Refs. 13 and 17 are visible in Fig. 1. Namely, the Ge-Ge peak has a greatly enhanced intensity around $E_L = 2.41$ eV (while the Si-Si vibration does not) and the Si-Si peak starts increasing its intensity as photon energy increases beyond that point (while the Ge-Ge peak does not). This points to the existence of electronic transitions within the Ge layers for photon energies in the range of $2.4 - 2.5$ eV, and others which are confined in the Si layers at higher photon energies.[13,17] It also means that the Ge-Ge (Si-Si) LO vibration does not propagate in the Si (Ge) layer. For the Ge-Ge vibration this is not obvious, since the bulk Ge-LO dispersion relation has a small overlap with the Si longitudinal acoustic dispersion and the former could propagate, with very small amplitude, into the Si layer.[5–7] However, resonant Raman data[9,13,17,18] show that this penetration is negligible, and the results of Fig. 1 also reflect this fact. More importantly for the discussion in hand, the spectra displayed in Fig. 1 calls attention to a different aspect of the laser-wavelength-dependence of the Raman spectrum. Namely, that the line shapes of the Ge-Ge and the Si-Ge modes suffer great changes as λ_L is varied. These changes in position and line shape for the 6QW sample occur for laser photon energies around the resonant peak in the Ge-Ge line. In contrast, the line shape and positions of these lines in the 2QW and 1QW samples are insensitive to the variations of laser frequency. Since the aforementioned resonance moves towards much higher energies in these two samples,[13,17] we associate the changes observed in the 6QW spectrum with resonant effects and we shall relate these effects to terracing at the Ge/Si interfaces. In order to approach this subject in a systematic manner, we start by comparing the spectra of the three different samples. Because of the resonant effects already mentioned, this comparison is only meaningful when made between spectra taken at frequencies that are off resonance. This is true for the $E_L = 1.96$-eV ($\lambda_L = 633$ nm) line in the case of the 6QW sample, and for any of the laser lines used in these experiments for the other two samples. On the other hand, spectra taken with $\lambda_L = 633$ nm are much weaker and have a poorer signal to noise ratio, especially for the 1QW and 2QW samples. Hence, for these two samples we use spectra taken with $\lambda_L = 633$ nm whenever possible and with $\lambda_L = 514.5$ nm when the ones taken with the previous line are too noisy. The forthcoming analysis is to be carried out having two premises, or propositions, in mind:

(i) Two different types of interfaces are incorporated in our samples, one is that between the Ge well and the 300-Å-wide Si spacer layers and the other is between the Ge well and the 5-ML Si barrier. The quality of the first is considerably better with respect to both the homogeneity of the alloy layer and the uniformity of the border between this and the Ge layer.

(ii) Both the TO and LO Ge-Ge modes decay within the interface region and none propagates into a Si layer. This is assumed for the two types of interface of proposition (i). Proposition (ii) is independent of proposition (i), although the extent of penetration into the alloy evidently depends on the details of the alloy composition. Proposition (i) seems sensible. The first type of interface is supported by a thick layer of the substrate material and should be of higher quality, especially for strained quantum wells. The 1QW sample contains only that type of interface, both types are equally present in 2QW sample, while in the 6QW sample the second type of interface dominates.

We begin our discussion with the Ge-Si mode, since this mode is related to the interface and should be localized around it.[6,7] In Figs. 2(a) and 2(b), we compare the off-resonance spectra of this mode in the three samples, for L and T polarizations, respectively. It is clear that the line corresponding to the Ge-Si mode is much wider in the 6QW sample than in the other two samples. This effect is only very weakly observed in the main peak of the TO spectrum. In the calculations for these modes performed by de Gironcoli *et al.*,[6,7] 2 ML of $Si_{0.5}Ge_{0.5}$ separate the Ge from the Si region. Both the composition of this layer and the thickness of all layers are assumed to be constant. Within this model, the LO mode is localized on the atomic plane of the alloy interfacial layer closest to the Ge quantum well, while the TO mode involves both alloy layers and spills over on both sides into the first monolayer of the pure Si and Ge regions

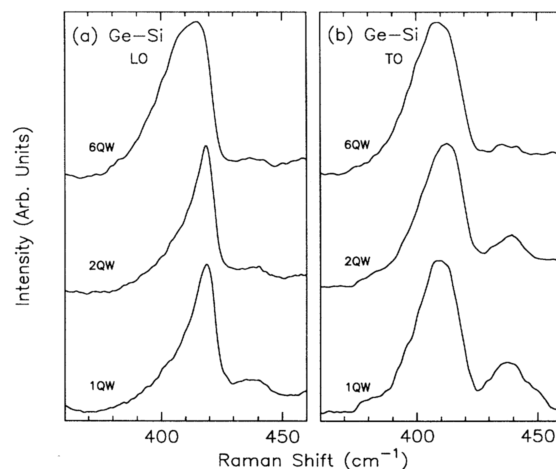

FIG. 2. Raman spectra for LO (a) and TO (b) Ge-Si modes of the three samples, at laser excitation $E_L = 2.41$ eV.

(see Fig. 3 in Ref. 6). If the assumption of homogeneity (both in atomic molar fraction and width) were relaxed, then contributions from regions of different widths and/or compositions should get different weight from the various Ge-Si clusters[16] and the line would be wider than that arising from a uniform interfacial alloy layer. Because of its greater localization, the LO line should be much more sensitive to this effect. Since the 6QW sample has the least perfect interface [proposition (i)] then it should also have the wider Ge-Si line, as indeed Fig. 2(a) shows.[19] Hence, the off-resonance behavior of the Ge-Si LO line gives a strong indication that the interfacial alloy layer is not homogeneous in width and/or alloy composition. The TO line, on the other hand, is less sensitive to these details, because it extends on both sides of the alloy layer. However, this line has a complex structure composed of a main line, for which the above assertion is true, and satellite lines of much smaller intensities at the higher energy side of the spectrum [Fig. 2(b)]. According to de Gironcoli and Baroni,[16] these satellite lines come from clusters that are richer in Si.[16] In our samples, these satellite lines are clearly visible in the 1QW sample and become progressively more blurred as we go to the 2QW and 6QW samples (Fig. 2). This blurring could be the result of averaging over a larger variety of types and sizes of clusters, thus arguing in favor of the progressively greater inhomogeneity of the interface layers as one progresses from 1QW to 2QW and 6QW samples.

In Fig. 3 we show the off-resonance Raman spectra of the three samples, for L [Fig. 3(a)] and T polarizations [Fig. 3(b)] in the frequency region of the Ge-Ge vibration. The spectra show a variation of line shape (asymmetry and linewidth) and peak position for the different samples. As before, this difference is more marked for the LO modes than for the TO ones. The peak positions of each line move systematically from higher to lower energies (see vertical dotted lines in Fig. 3) as we progress from 6QW to 2QW and 1QW samples. This might be naively attributed to tunneling through the Si barrier

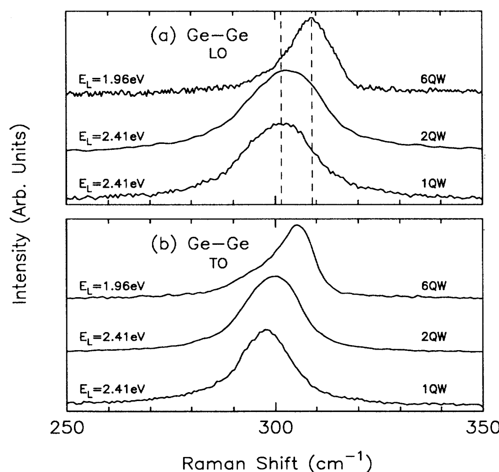

FIG. 3. The off-resonance Raman spectra for LO (a) and TO (b) Ge-Ge modes of the three samples. The dashed lines are intended as a guide to the eye.

into the neighboring quantum wells, which would result in a similar effect. While this is theoretically possibly for the LO mode, it is not so in the case of the TO vibrations. So, if this were the correct explanation, the LO line should shift to higher energies as the number of quantum wells increases (less effective confinement) while the TO line should exhibit the same position in all three samples, being always confined within a *single* Ge quantum well. This is clearly not the case, as shown in Fig. 3(b). Also, this would contradict the resonant Raman results,[13,17] which argue in favor of the hypothesis made in proposition (ii), where the Ge-Ge LO phonon does not penetrate into Si layers. So let us stick to our proposition (ii), which asserts that both the TO and LO Ge-Ge modes decay into the interface alloy layers, but do not actually tunnel into the Si layer. The calculations of Ref. 6 show that the LO vibration decays faster into the interface layer than the TO one (Fig. 3 of Ref. 6). This can also be seen qualitatively by comparing the dispersion relation for optical phonons of bulk Ge and $Si_{0.5}Ge_{0.5}$ (see Fig. 1 of Ref. 9). Although the former starts at a higher energy at $\vec{q}=0$, especially when strain is taken into account, the decrease in frequency of the bulk Ge imposed by confinement would cause the confined mode (equivalent to a $\vec{q}\neq 0$ bulk Ge mode) to overlap with some part of the alloy dispersion relation. Since the decrease in frequency for finite $\vec{q}$ is steeper for the TO than for the LO phonon, the former begins to overlap with the alloy dispersion relation at lower $\vec{q}$ (less confinement, or larger effective confinement width) than the LO phonons. Thus, the effective confinement width of the TO phonons is larger than that of the LO phonons, i.e., the TO mode penetrates deeper into the alloy layer. A similar result for the Si-Si mode was found experimentally by Schorer *et al.*[9]

The results of off-resonance spectra of the three samples (Fig. 3) are now understandable in terms of our two propositions. The interfaces between the Ge-Si layers improve in smoothness and uniformity as we progress from the 6QW to the 2QW and the 1QW samples. If the greater "roughness" in the multiple quantum wells includes a distribution of effective confinement lengths (terracing), the observed line shape is composed of several peaks with a predominance in the spectrum (volume effect) of the peaks originating in the regions of larger well width (higher frequencies). Thus, the line is more asymmetric in the 6QW sample and its peak is shifted toward higher frequencies than those of the 2QW and 1QW samples. The latter two samples have smoother interfaces with a narrower distribution of effective well widths. The modes in these two samples are more confined, their dispersion relations are steeper, and even small fluctuations in well width produce a broadening of the line, without much affecting its intensity. Thus, the line shape for 2QW and 1QW samples becomes symmetrical, broader, and peaks at lower energies than that of the 6QW sample. These are indeed the results shown in Fig. 3. This terracing affects the LO modes more drastically than the TO mode, because the latter penetrates deeper into the alloy layer and is, therefore, less confined. This results in a narrower line shape for TO modes than for LO modes.

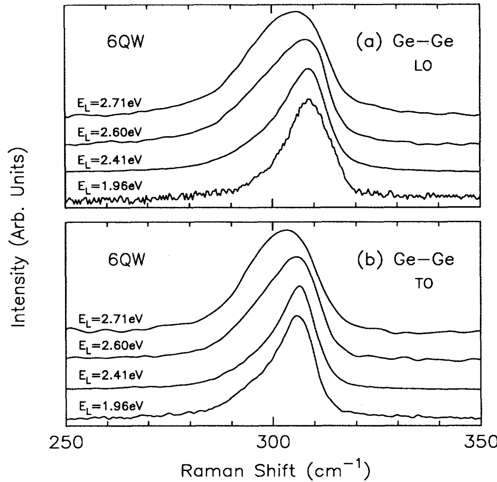

FIG. 4. The Ge-Ge Raman line in the 6QW sample for different exciting laser photon energies, LO modes (a) and TO modes (b).

This difference is clear [Figs. 3(a) and 3(b)] in the spectra of the 1QW and 2QW samples, whereas it hardly shows in the 6QW sample, on account of the wider distribution of confinement lengths in this sample. Because of the different effects of terracing on TO and LO lines care must be taken when unfolding these superlattice modes onto the bulk dispersion relation.

Finally, we turn our attention to the unusual changes in shape of the Ge-Ge Raman line in the 6QW sample as the laser energy increases above the 2.41 eV (Fig. 4). As we remarked earlier, around this frequency there is a resonance produced by electronic transitions confined in the Ge layers.[13,17] In the range between $E_L = 1.96$ eV and $E_L = 2.41$ eV the position and shape of the line remains approximately constant, although the overall intensity of the line increases sharply as $E_L = 2.41$ eV is approached. However, as the laser excitation energy gets higher, the peak gradually broadens and its center of mass shifts towards lower frequencies. Thus, the line, which is highly asymmetric for $E_L \leq 2.41$ eV, transforms into a more symmetric and significantly broader band for larger E_L's. The effect is rather impressive and is not observed in either the 2QW or the 1QW samples.

According to our previous reasoning, the main difference between the 6QW sample and the other two samples is that in the former there is a distribution of well widths due to large-scale roughness. This distribution is the main cause for the high asymmetry of the line away from resonance, as was already discussed. The asymmetric line shape does not reflect in a simple manner the width distribution, because the weight of larger widths (higher frequencies) is larger and contributes with a higher intensity. As the laser photon energy increases, resonant conditions are met for electronic states confined in locally narrower wells, and thus LO intensity contributed from such local wells is enhanced. These LO phonons have lower frequencies the narrower is the well. Since a distribution of such widths exists, by changing laser energy a continuous shift of the peak position is observed together with a marked change in line shape, broadening towards lower frequencies. Notice that in order to have this selective resonance, the regions of a given width have to be of a size comparable to the coherence length of the states involved in the electronic transitions (exciton radius or some similar parameter). This means that the type of interface roughness responsible for this effect is one on the 10^2-Å scale, i.e., this selective resonance indicates the existence of terracing of a similar nature as that occurring in III-V materials.[1,10] The TO line is also resonanting, though not to the same extent, and the contributions to its lower frequency tail come from sites less confined and therefore the effect is less impressive, but they are still evident in Fig. 4(b).

Should 1QW and 2QW samples show similar effects? We do not get close enough to resonance for those samples. The higher resonance energy, in itself, can also be understood in accordance with our two propositions, namely, higher confinement of the electronic states as well and therefore higher energies. This means that the effect is not likely to be as impressive as in 6QW sample and if at all, should be more noticable for 2QW sample. A *much* smaller effect is indeed seen also in the case of 2QW sample (not shown).[20] We do believe that the picture of the Ge/Si interface, proposed in this work, fully explains all our data. One can also infer that resonance measurements of Ge/Si superlattices are heavily dependent on the interface, which is affected by the number of layers in the barrier and, of course, by the growth parameters, which were not dealt with here.

In our discussion we neglected the higher orders of the Ge-Ge confined mode for the following reasons: their frequencies are expected to be lower than the range of frequencies we observed and also their intensity should be much lower. Moreover, such phonons, if observed, are not so confined and therefore significantly narrower.

In conclusion, the detailed line-shape study of the Raman spectrum of our samples can be self-consistently explained by invoking both small-scale roughness (alloying at the interface) and large-scale thickness variations of the layers (terraces). In particular, the effects of terracing are very pronounced near resonance. Hence, the analysis of resonant Raman data of Ge/Si multiple quantum wells and superlattices, even those with the *best* available interfaces, should always be interpreted in the light of the effects of terracing on the observed intensities and line shapes.

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[19] Although the line shape of the Ge-Si LO vibration for the 6QW sample shows small variations as λ_L changes, it is always much wider than the corresponding line in the other two samples.

[20] The absence of this effect in these two samples proves that the selective resonant Raman effect originates in a width distribution within *each* quantum well rather than in width fluctuations from one well to another, since all three samples have many repeated quantum wells fabricated in identical conditions.