Quantum Dots: Phonons in Self-Assembled Multiple Germanium Structures

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INTRODUCTION

Self-assembled Ge quantum dots by the Stranski–Krastanov growth mode have attracted much attention for many years. Similar to the purpose of the research on short-period Si/Ge superlattices and Er-doped Si, self-assembled Ge/Si quantum dots may be exploited to fabricate Si-based on-chip light emitting sources for 1.55-μm fiber optic communication applications. Takagahara and Takeda[1] and Ren[2] have theoretically predicted that an indirect-to-direct conversion of the optical transition of SiGe quantum dots would occur whenever the sizes of the quantum dots were small enough. In order to increase the component of quasi-direct transition of the Ge/Si quantum dot system and thus to enhance light emission intensity of the quantum dots for practical applications, the understanding of nonradiative recombination mechanisms (such as phonon-assisted process) is essential.

To probe phonons from self-assembled semiconductor quantum dots, Raman spectroscopy is an efficient and indispensable experimental tool. Up to date, several groups have already reported Raman scattering studies of self-assembled quantum dots of group III–V systems, such as (In, Ga, Al)Sb/GaAs,[3] InSb/InP,[4] In(Ga)As/GaAs,[5–9] InAs/InP,[10–13] (Al, Ga)As/InAs,[14] InAs/AlAs,[15,16] and GaN/(Si)AlGaN,[17,18] group II–VI systems, such as CdSe/ZnSe,[19] and group IV system, i.e., Ge/Si.[20–38] Optical phonon spectra of any of these systems were basically used to extract the chemical composition of the quantum dots as a result of interdiffusion between the dots and the surrounding media or the substrates. It should be pointed out that a few other techniques for the determination of composition of self-assembled quantum dots have been reported. These include scanning tunneling microscopy,[39] transmission electron microscopy (TEM) with high-resolution imaging,[40] electron energy loss spectrometry,[41] X-ray energy dispersive spectrometry,[42] high-resolution X-ray diffraction,[43,44] and scanning TEM.[45] Most of these techniques are capable to show nonuniform dot material distribution in the dots. Optical phonon Raman scattering method is simple and direct to give an average concentration of the dots. In contrast to the tremendous research on optical phonons, the effort on the research of phonon process in the low-frequency acoustic spectral region, however, is much smaller. In self-assembled Ge quantum dot system, for example, one early work reported the observation of equal-distance acoustic peaks in 25-period Ge quantum dot superlattices.[21] Afterwards, Milekhin et al. investigated folded longitudinal acoustic phonons in their Ge dot superlattices and explained the acoustic vibrations by the elastic continuum model.[26–29] Recently, resonant Raman scattering by acoustic phonons in double- and multilayered Ge dot structures was reported and the observed equal-distance oscillation peaks were explained by interference and ordering effects.[32,33] The origins of these observed low-frequency acoustic phonon spectra in Ge dot superlattices therefore remain unclear and debatable because there is a lack of systematic studies, such as the dependence of acoustic phonons on island sizes and other island-related parameters.[23]

In this entry, we systematically study Raman scattering by optical and acoustic phonons in multiple Ge quantum dots. The analysis of GeGe and SiGe optical phonon features takes the phonon confinement effect, strain effect, and atomic intermixing into account. Acoustic phonons are found to originate from folded acoustic phonons associated with a superlattice and can be explained by elastic continuum model.

RESULTS ON SELF-ASSEMBLED MULTIPLE GE QUANTUM DOTS

All samples were grown on Si (100) wafers using a solid source molecular beam epitaxy (MBE) system. Samples A, B, and C were grown at 540°C with 10 periods of Ge and Si bilayers and contained a Ge coverage of 12, 15, and
18 Å, respectively. Samples D and E were grown with the same Ge layer thickness of 15 Å but at different growth temperatures of 500°C and 600°C, respectively. In addition, sample D has 10-period Ge and Si bilayers while sample E contains 22 periods. The Si spacer layer thickness of 20 nm was used for all superlattices. No cap Si layers were grown intentionally on these samples for convenience of atomic force microscopy (AFM) characterizations. Transmission electron microscopy measurements were exploited as well to obtain the size, density, and uniformity of the self-assembled dots. Raman scattering measurements were performed on a Renishaw Raman Imaging 2000 system with a 514 Ar⁺ laser as the excitation light source in the backscattering configuration.

Fig. 1a shows a typical cross-sectional TEM image of sample A. Ten-period vertically correlated layers of Ge dots are evident. The dots in the different layers (beyond the third Ge layer) are identical in size and shape. This observation is also seen for other dot samples. Fig. 1b and c shows an AFM image, and height and base statistic analysis of sample A, respectively. Gaussian distributions were used to fit the data. Most of the dots appear as pyramid and the average dot base and height are 11.9 and 110.4 nm, respectively. Similar measurements have been performed on other samples as well. The growth parameters and structural data were summarized in Table 1. It should be noted that the structural data were from AFM measurements only and not calibrated by TEM characterizations.

**Optical Phonons**

Fig. 2 shows the Raman spectra of the dot samples in the spectral region of SiGe and GeGe optical phonons. Similar SiGe (from 400 to 420 cm⁻¹) and GeGe lines (near 300 cm⁻¹) are observed for dot samples. The frequency position of SiGe optical phonons more or less represents the degree of the interdiffusion between Si spacers and Ge dots. Samples A, B, and C have almost the same Si–Ge optical phonon line shape. The peak frequency changes slightly from 418 cm⁻¹ for sample A with a nominal Ge thickness of 1.2 nm to 416 cm⁻¹ for sample C with a nominal Ge thickness of 1.8 nm. To understand this, we shall notice that the interdiffusion arises from both thermal diffusion and strain-induced diffusion. As the same growth temperature of 540°C was used for the three samples, the degree of interdiffusion due to thermal management is the same. The slight difference in frequency therefore comes from the difference in strain-induced diffusion. The thicker Ge was deposited the more relaxation was presented in the dots (as will be discussed in detail in the following). Strain-induced interdiffusion in the samples with thicker nominal Ge thickness is smaller. The resulting lower frequency can be understood with the
following picture: The eigen-frequency of these optical phonons is determined by the effective stretching force constant for each bond and the mass of atoms at the end of each bond. For example, SiSi optical phonon band is centered at 520 cm \(^{-1}\) for all of the samples and the substrate (not shown in the figure), considering the force constant of SiSi to be 39.5 N/m, and atomic weight of 28.1 for Si.\(^{[46]}\) Likewise, optical phonons of crystal Ge can be found at 300 cm \(^{-1}\) with the GeGe bond force constant of 35 N/m and atomic weight of 72.6. Any mixture of Si and Ge leads to SiGe optical phonons with the frequency in between 300 and 520 cm \(^{-1}\). For simplicity, we assume that Ge quantum dots are fixed and only Si atoms move. At a situation of stronger interdiffusion, more Si atoms are introduced into Ge dots. Thus SiGe optical phonon band has a frequency closer to SiSi optical phonon band, which is exactly the case for samples A through C. Likewise, much lower frequency position of 406 cm \(^{-1}\) for sample D and slightly larger frequency of about 420 cm \(^{-1}\) for sample E compared with that of sample B (due to the same nominal Ge) can also be explained by the above picture.

Now let us analyze the GeGe optical phonons in the samples. It is important to note that the GeGe modes from the dot samples are different from the second-order transverse acoustic (2TA) phonon mode\(^{[47]}\) for Si at 303 cm \(^{-1}\). For comparison, the vertical dotted line is plotted at 300 cm \(^{-1}\) to represent the optical phonon position for bulk crystalline Ge. The frequency positions of the GeGe optical phonons in dot samples are shifted slightly to higher frequencies with respect to their bulk value. There are several mechanisms to cause a Raman shift of GeGe optical phonons. The first one is phonon confinement. It is known that the optical phonon branches of bulk Ge are quadratic and, moreover, nearly flat at the Brillouin zone center \((k\approx0)\). Confined optical phonons in a nanocrystal are equivalent to those vibrations in an infinite crystal whose wave vector is given by \(m\pi/d\), where \(m\) is an integer and \(d\) is the size of the nanocrystal, in our case, the height of the dots. In the dots with a very small height of 1.5–2 nm,\(^{[21]}\) the phonon confinement effect was observed to result in a shift less than 2 cm \(^{-1}\). As the dot size for the present samples is much larger than the lattice constant of Ge, the wave vector is extremely small, leading to insignificant phonon confinement effect.

The size confinement effect of optical phonons in quantum dots with small size should give rise to a shift of phonon band to lower frequency side due to the negative dispersion of the optical phonon branch. A compressive strain on the dots in the lateral directions as a result of the lattice mismatch of Si and Ge, however, leads to a GeGe mode shift to the higher frequency side. This concept can be written as:\(^{[48]}\)

\[
\omega = \omega_0 + \frac{1}{2\omega_0} [p\epsilon_{zz} + q(\epsilon_{xx} + \epsilon_{yy})]
\]

where \(\omega\) is the Ge-Ge mode frequency induced by a biaxial strain, \(\omega_0\) is the frequency of the Ge zone-center LO phonon; \(p\) and \(q\) are the Ge deformation potentials; \(\epsilon_{xx}\) and \(\epsilon_{yy}\) are the biaxial strain \((a-a_0)/a\) with \(a\) and \(a_0\) being unstrained and strained lattice constants, respectively. The physical parameters used in Eq. 1 can be obtained from

<table>
<thead>
<tr>
<th>Sample</th>
<th>Ge layer thickness (Å)</th>
<th>Si layer thickness (nm)</th>
<th>Growth T (°C)</th>
<th>Dot base (nm)</th>
<th>Dot height (nm)</th>
<th>Density (cm(^{-2}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>12</td>
<td>20</td>
<td>540</td>
<td>110.4</td>
<td>11.9</td>
<td>3.6 \times 10^9</td>
</tr>
<tr>
<td>B</td>
<td>15</td>
<td>20</td>
<td>540</td>
<td>122.0</td>
<td>14.0</td>
<td>4.1 \times 10^7</td>
</tr>
<tr>
<td>C</td>
<td>18</td>
<td>20</td>
<td>540</td>
<td>122.2</td>
<td>16.0</td>
<td>3.5 \times 10^9</td>
</tr>
<tr>
<td>D</td>
<td>15</td>
<td>20</td>
<td>500</td>
<td>114.7</td>
<td>15.1</td>
<td>5.9 \times 10^8</td>
</tr>
<tr>
<td>E</td>
<td>15</td>
<td>20</td>
<td>600</td>
<td>175.5</td>
<td>10.2</td>
<td>2.6 \times 10^8</td>
</tr>
</tbody>
</table>

Fig. 2 Raman spectra of the dot samples. Similar SiGe and GeGe lines are observed for these samples. The frequency positions of the GeGe optical phonons in dot samples are shifted slightly to higher frequencies with respect to their bulk value (300 cm \(^{-1}\)).
Ref. [34]. For fully strained pure Ge on Si, we obtain \( \omega = 317.4 \text{ cm}^{-1} \). The large difference between this calculated number and the experimental GeGe mode values (close to 300 cm\(^{-1}\) as shown in Fig. 2) suggests that the dots are not fully strained as assumed in the calculation. By using Eq. 1 and the experimental GeGe optical phonon frequencies \( \omega_a \), the residual biaxial strain \( (\varepsilon_{xx}) \) on the quantum dots can be estimated and listed in Table 2. Very small residual strain values obtained suggest the dots are almost fully relaxed.

An obvious reason of the strain relaxation is the atomic intermixing at the Si–Ge interface. The degree of the intermixing at the Si–Ge interface. The degree of the interface intermixing can be determined by the integrated peak intensity ratios, and indicate that the different GeGe mode frequency is \( I_{\text{GeV}}/I_{\text{SiGe}} \) as shown in Fig. 2, leading to a GeGe mode frequency of 308.3 cm\(^{-1}\). The calculated values show a considerable shift from the value obtained for fully strained Ge on Si (317.4 cm\(^{-1}\)). However, these are still larger than the peak frequencies of the GeGe modes of the samples A through E. As a matter of fact, the absolute value of the residual strain on the dots shown in Table 2 is much smaller than the absolute value estimated here by interdiffusion only.

Table 2 The experimental GeGe mode frequencies, integrated peak intensity ratios, and calculated Ge composition in dots and residual in-plane strain in the dot samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>( \omega(\text{GeGe}) ) (cm(^{-1}))</th>
<th>( \omega(\text{SiGe}) ) (cm(^{-1}))</th>
<th>( I_{\text{GeV}}/I_{\text{SiGe}} )</th>
<th>Ge composition in dots</th>
<th>( \varepsilon_{xx} ) ((10^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>302</td>
<td>418</td>
<td>1.45</td>
<td>0.48</td>
<td>−4.8</td>
</tr>
<tr>
<td>B</td>
<td>301.1</td>
<td>417.5</td>
<td>1.45</td>
<td>0.48</td>
<td>−2.65</td>
</tr>
<tr>
<td>C</td>
<td>300.5</td>
<td>416</td>
<td>1.45</td>
<td>0.48</td>
<td>−1.2</td>
</tr>
<tr>
<td>D</td>
<td>302.4</td>
<td>406</td>
<td>2.14</td>
<td>0.58</td>
<td>−5.8</td>
</tr>
<tr>
<td>E</td>
<td>300.4</td>
<td>419.5</td>
<td>1.29</td>
<td>0.45</td>
<td>−0.96</td>
</tr>
</tbody>
</table>

The experimental GeGe mode frequencies, integrated peak intensity ratios, and calculated Ge composition in dots and residual in-plane strain in the dot samples.

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\[
I_{\text{GeV}}/I_{\text{SiGe}} = B \frac{x}{2(1-x)}
\]

where \( x \) is the average Ge concentration and the coefficient \( B \) is related to the Bose factor and the frequencies of GeGe and SiGe optical modes of the alloy. It is found that the coefficient \( B \) varies weakly with alloy composition and is determined to be 3.2. \( I_{\text{GeV}}/I_{\text{SiGe}} \) can be obtained from the spectra in Fig. 2. The Ge concentrations in quantum dots estimated by this method are listed in Table 2. It should be noted that the estimation by such interpolation is rough because the Si 2TA phonons and optical phonons from wetting layers have not been subtracted in the calculation. Moreover, the composition obtained is an average value, which does not show any information about composition distribution in the dots.

Fig. 3 plots the GeGe peak position as a function of the Ge concentration in the dots. Solid straight lines 1 and 2 are used to guide the eyes and indicate that the different GeGe mode frequency is obtained for dots not only with different Ge concentrations but also with a similar Ge concentration. Solid line 3 is a calculation result using Eq. 1. In this calculation, we assume the same \( \omega_0 \) and other deformation potentials for pure Ge. This approximation is reasonable because the interdiffusion occurs in the interface and the “core” of the dots remains pure Ge. For example, the nonuniform Ge content in the dots has been proved by elemental distribution analysis using an electron energy filtering imaging method in TEM and scanning TEM. The in-plane strain \( \varepsilon_{xx} \) used can be written as \( (\alpha_{\text{Si}} - \alpha_{\text{Ge}})/\alpha_{\text{Si}} \), with the lattice parameter of an alloy with Ge concentration of \( x \) determined by Vegard’s law \( \alpha_{\text{Si}} = x\alpha_{\text{Si}} + (1-x)\alpha_{\text{Si}} \), where \( \alpha_{\text{Ge}} \) and \( \alpha_{\text{Si}} \) are lattice parameters of pure Ge and Si, respectively. Simple calculations show, for example, in-plane strain to be \( −0.02 \) for an alloy with Ge concentration of 0.48 (samples A, B, and C), leading to a GeGe mode frequency of 308.3 cm\(^{-1}\). The calculated values show a considerable shift from the value obtained for fully strained Ge on Si (317.4 cm\(^{-1}\)); however, these are still larger than the peak frequencies of the GeGe modes of the samples A through E. As a matter of fact, the absolute value of the residual strain on the dots shown in Table 2 is much smaller than the absolute value estimated here by interdiffusion only.
Moreover, the picture of interdiffusion does not completely account for the peak position difference for the different samples with the same Ge composition as of line 3 in Fig. 3. The above analysis suggests that additional strain relaxation mechanisms exist apart from Ge/Si interdiffusion. We believe that this additional strain relaxation comes from the process of the formation of Ge dots.

The relaxation of the dots in addition to atomic interdiffusion consists of the relaxation of the surface-layered dots and the relaxation of the embedded dots. The relaxation of the surface-layered dots is related to the nature of Stranski–Krastanov growth mode. When Ge is deposited on Si, it first takes a layer-by-layer growth format. As the Ge film exceeds its critical thickness, it becomes rough by forming pyramidal islands to relieve strain energy. As more Ge is deposited, the islands transform from the pyramid shape to the dome shape, leading to more strain relaxation. Atomic force microscopy measurements on our samples indeed showed, for example, the pyramid-over-dome ratio decreases as the increase of the Ge deposition for these samples grown at the same temperature of 540°C.[25] The relaxation is also induced by the dot embedding effect. Sutter and Lagally have used a strain-driven atom diffusion model to show that the dot shape transforms to truncated shape after the growth of Si layer on top.[52] Indeed, we have observed this transition from the cross-section TEM measurements.

Because of the capping process, the in-plane strain is relaxed.[39]

After recognizing all the above relaxation mechanisms, we can fully explain the observed GeGe optical phonon frequencies. First, let us take a look at line 1 of Fig. 3. This line represents the GeGe mode trend of a set of samples (C, D, and E) grown at different temperatures. GeGe mode frequency changes from 302.4 to 300.4 cm\(^{-1}\) when the Ge concentration decreases from 0.58 to 0.45 as a result of growth temperature from 500°C to 600°C, indicating that the sample grown at higher temperature induces stronger interface intermixing, or more strain relaxation. Quantitative calculations using Eq. 1 show that a shift of 2.2 cm\(^{-1}\) is obtained for the present interdiffusion difference in the three samples (as can be seen from line 3 in Fig. 3). This number is almost the same as the experimental observed shift, suggesting that the strain relaxation from the different degrees of interdiffusion is the main factor responsible for the different frequencies of the GeGe mode in this set of samples. In other words, the overall effect of the dot shape transition on the GeGe mode frequency is about the same for the three samples. Line 2 in Fig. 3 represents another set of samples (A, B, and C). As shown previously, this set of samples has the same Ge concentration in the dots. Nevertheless, the GeGe optical phonon frequency is different and the largest difference is 1.5 cm\(^{-1}\). This suggests different strains are present on the dots in different samples, which arises from the different dot morphologies in the different samples, such as pyramid-over-dome ratios, dot height-over-base ratios, and embedding effect.

The above-analyzed, negligible phonon confinement effect but dominant atomic intermixing and/or strain effect in determining frequencies of optical phonons for quantum dot samples can also be proved by performing annealing experiments. Fig. 4a shows the Raman spectra of sample A under rapid thermal annealing (RTA) for 5 min at different temperatures. The vertical dotted line is plotted at the center of GeGe mode from as-grown sample for comparison. The GeGe Raman line broadens and shifts to lower frequencies as the annealing temperature increases. This phenomenon was also observed when annealing other samples. Fig. 4b shows the annealing temperature dependence of the Raman shift for the dot samples grown at different temperature. After 850°C annealing for 5 min, the GeGe optical mode shifts downward by as much as 8, 6–7, and 5 cm\(^{-1}\) for the samples grown at 500°C, 540°C, and 600°C, respectively. The intermixing after annealing leads to fully relaxed SiGe alloy-like dots, causing the observed peak shift.

![Fig. 4](image-url)
Acoustic Phonons

Fig. 5a shows the Raman spectra of the samples A, B, and C, which were grown with the same temperature and different Ge layer thickness. The vertical dot lines are used to facilitate the comparison among the samples. Similar SiSi, SiGe, and GeGe optical vibration peaks are seen at around 520, 415, and 300 cm\(^{-1}\), respectively. Besides these peaks, low-frequency oscillation peaks are observed for all of the three samples around 84.3, 104.7, 126.6, 150, and 171.8 cm\(^{-1}\), respectively. These nearly equal-distance peaks with a period of about 20 cm\(^{-1}\) are higher-order acoustic phonons. The lower orders were attenuated by the Rayleigh line rejection filter and thus were not observed. An interesting phenomenon is that the acoustic peak frequencies do not scale with the dot size. Lamb’s theory predicts that confined phonon frequency should be inversely proportional to the nano-crystal size\(^{[53]}\) and was shown to be true for a number of nano-crystal systems such as CdS\(_x\)Se\(_{1-x}\) in glasses\(^{[54]}\) and Si in SiO\(_2\) matrix.\(^{[55]}\) The present result suggests that the observed low-frequency peaks are not confined phonon modes in quantum dots.

Fig. 5b shows the Raman spectra of the samples A, D, and E, which were grown at different temperatures. The vertical dot lines are again used here to facilitate the comparison among the samples. Acoustic phonons for different samples have different frequencies. Specifically, acoustic phonon peak frequencies for sample A have been assigned in Fig. 5a. Sample D has acoustic phonons around 71.4, 95.7, 118.8, and 141 cm\(^{-1}\), and sample E has acoustic phonons around 83.8, 105.8, 126.8, and 149.8 cm\(^{-1}\). The period of these nearly equal-distance peaks is about 24, 22, and 20 for samples D, E, and A, respectively.

There are two possibilities, which may be used to explain the observed periodic acoustic phonons.\(^{[56]}\) The first one is that these are probably phonon modes confined in quantum wells where zone-edge phonon modes have been made, “allowed” by breakdown of wave vector conservation. The second one is that they are associated with a superlattice where low-frequency zone-edge phonon modes have been folded into zone center. The first concept has been used to explain the periodic acoustic phonon oscillations in the InAs/InP self-assembled quantum dots.\(^{[12]}\) The period of the periodic acoustic phonons can be expressed by:

$$\Delta \omega = \frac{1}{2L} \cdot \frac{v}{c}$$

where \(L\) is the thickness of the quantum layer, \(v\) is the sound velocity of the phonons, and \(c\) is the speed of the light. If the observed phonons were the confined phonons in the Si spacer layers, then with the velocity \(v_{Si} = 8.44 \times 10^5\) cm/sec\(^{[57]}\) and the peak periods of 20–24 cm\(^{-1}\) for the present samples, we would obtain the Si layer thicknesses for our samples to be around 5.9–7 nm. The designed thickness of Si spacer thickness for all the samples was 19–20 nm. The actual effective thickness of Si spacer layers has shrunk to around 14 nm characterized by cross-sectional TEM. The reason is that Si spacers were deposited on curvature surfaces containing three-dimensional dots. A similar situation was often observed when growing short-period superlattices on relaxed virtue substrates, which consisted of cross-hatch undulation patterns. The layer deposited on top was stretched as a result of undulation and the effective thickness along the growth direction was shrunk. The effective thickness of about 14 nm for the present samples, however, is still more than two times of the calculated 5.9–7 nm, suggesting that these phonons have nothing to do with the confinement in the Si spacers. Likewise, the phonons are not confined phonons in the wetting layers either. Samples grown at different temperatures lead to different
wetting layer thickness because of Si/Ge interdiffusion. Different from the dots grown at much lower temperature (300°C)\textsuperscript{26,29} or by the use of antimony as surfactant,\textsuperscript{34} where the wetting layers were more or less around 3–4 MLs, the wetting layers in this set of samples had the thickness in the region of 15–20 Å with cross-sectional TEM characterizations (not shown here). Therefore the wetting layers are not pure Ge but SiGe alloy. Phonon velocity for SiGe can be obtained by the linear interpolation of LA phonon velocity of Si and Ge. Simple calculations showed that one could never obtain an agreeable solution compared with the experimental results.

The above analysis leaves the only possible origin for these phonons, i.e., they are folded acoustic modes related to the periodicity of the superlattice. Rytov’s elastic continuum model\textsuperscript{58} has been well applied for folded acoustic phonons in quantum well superlattices and was also seen to explain the acoustic phonon features in Ge dot multilayers\textsuperscript{26–29} and InAs/GaAs dot multilayers.\textsuperscript{14} In this model, the acoustic phonon dispersion can be written as:

\[
\cos(qd) = \cos\left(\frac{od_1}{V_1}\right) \cos\left(\frac{od_2}{V_2}\right) - \frac{1}{2} \left( \frac{R + \frac{1}{R}}{V_1 + V_2} \right) \times \sin\left(\frac{od_1}{V_1}\right) \sin\left(\frac{od_2}{V_2}\right)
\]  

(4)

where \( q \) is the superlattice wave vector perpendicular to the layers and is roughly determined to be \( q \approx \frac{4\pi n’}{\lambda_L} \), where \( \lambda_L \) is the incident laser light wavelength and \( n’ \) is the refractive index of the material at that wavelength. In addition, \( R = \frac{V_1\rho_1}{V_2\rho_2} \), \( d = d_1 + d_2 \), \( d_1 \) and \( d_2 \), \( \rho_1 \) and \( \rho_2 \), and \( V_1 \) and \( V_2 \) are the thickness, density, and sound velocity in Ge and Si layers, respectively. The physical parameters can be obtained from Ref. [57]. The model designed for quantum well superlattice can be considered as a zero-order approximation to the real quantum dot superlattice as it does not take into account quantum dot shape, size, and density. It was suggested in Ref. [27] to use nominal thickness of Ge layer as an effective thickness \( d_2 \). Nevertheless, with this rough approximation, good agreement with experimental data was achieved\textsuperscript{26–29} Following this approach, with the effective Si spacer layer thicknesses of about 14 nm and wetting layer thicknesses of about 1.5 nm in our samples, we calculated the phonon dispersion and showed acoustic phonon peak period of 19–21 cm\textsuperscript{-1}, which was close to the experimentally observed 20–24 cm\textsuperscript{-1}. Therefore we conclude that the observed low-frequency modes are folded acoustic phonons. More accurate description of the acoustic spectra of semiconductor quantum dot superlattices will require rigorous calculations of acoustic phonon scattering on quantum dot as well as multiple scattering effect produced by quantum dot ensemble in each layer.

**CONCLUSION**

We have studied Raman scattering from optical phonons and acoustic phonons in the Ge quantum dot superlattices. The GeGe optical phonon frequencies of the dot samples were larger than 300 cm\textsuperscript{-1} but within 3 cm\textsuperscript{-1}, indicating that the dots were relaxed in the superlattices. Strain relaxation was found not only from the atomic interdiffusion but also from the dot morphology transition both for the surface dots and the embedded dots. The annealing experiments also proved that the strain relaxation and intermixing were the key parameters for the observed optical phonons. Optical phonon studies show that Raman spectroscopy is an efficient tool for the determination of the average compositions and strains for zero-dimensional quantum dots. Periodic oscillations in the lower frequency region were observed for quantum dot superlattice samples. These acoustic phonons were found to be related to superlattice period and could be well explained by the elastic continuum model.

Furthermore, the method for analysis of phonons in the above Ge quantum dot superlattice system can be generally used for other quantum dot systems, such as, InGaSb/GaAs, InSb/InP, InGaAs/GaAs, InAs/InP, AlGaAs/InAs, InAs/AlAs, and GaN/(Si)AlGaN, CdSe/ ZnSe, and so on. Phonon spectra in these material systems depend on the physical parameters of superlattice, such as concentration in dots, dot size, and period thickness. In the case of very small dots with high density (e.g., greater than 10\textsuperscript{11} cm\textsuperscript{-2}), multiple scattering effects may play an important role in the modification of phonon spectra. The phonon densities of states in quantum dot superlattices change as seen from the strong modification of phonon spectra with the appearance of quantum dots and can be used to calculate the thermal conductivities of superlattices. Research has shown a great reduction of thermal conductivities in quantum dot superlattices, which have great applications in the fabrication of new-generation solid-state thermoelectric devices. Phonon spectra modification by quantum dots also suggests the potential applications of quantum dot superlattices as phonon lenses, filters, and reflectors for phonon-assisted lasers in indirect-band gap material systems.

**ACKNOWLEDGMENTS**

The authors wished to thank Dr. S. G. Thomas of Motorola Semiconductor Products Sector for performing some of the TEM characterizations and Prof. Peter Y. Yu of UC Berkeley for valuable discussions. This work was funded by the DoD MURI-ONR program on Thermoelectrics (N00014-97-1-0516) and the AFOSR MURI program on phonon engineering (F49620-00-1-0328).
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