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Strain and Phonon Confinement in Self-Assembled Ge Quantum Dot Superlattices *

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Raman scattering measurements were carried out in self-assembled Ge quantum dot superlattices grown by molecular beam epitaxy. The characteristics of the Ge-Ge, Si-Ge, Si-Si$_{\rm LOC}$ and Si-Si peaks were investigated, especially the Ge-Ge optical phonon frequency shift was emphasized, which was tuned by the phonon confinement and strain effects. The experimentally observed frequency shift values of the Ge-Ge peak frequency caused by optical phonon confinement and strain in Ge quantum dots were discussed with quantitative calculations.

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Self-assembled Ge quantum dots (QDs) have attracted growing interest because they show optical and electronic properties much different from bulk solid state materials. Both fundamental physical properties and potential applications in novel devices of Ge QDs have been widely investigated.[1] To study the phonon mechanism in Ge QDs is indispensable since phonons are very important for luminescence in indirect band gap semiconductors. In this Letter, we report on the investigation of optical phonon mode and its relation to the phonon confinement and strain in Ge quantum dot superlattices by Raman scattering measurements. Optical phonon confinement effects have been extensively investigated in two-dimensional (2D) semiconductor heterostructures and 1D systems. Owing to the difficulties of experimental observation caused by fluctuations of size, shape, and orientation in 0D systems, however, the confinement effects are not so much to be investigated. So far, only a few relative works have been reported on Si[2−4] and Ge.[5] In the present experiments, we found that the Ge-Ge peak in the Raman spectra are shifted slightly to their bulk value 300 cm^{-1}, which was deduced to be mainly attributed to the lateral compressive strain and phonon confinement in the dots. The main purpose is to understand the characteristics of the strain and the optical phonon confinement in self-assembled Ge quantum dot superlattices. The experimentally observed optical phonon frequency shift values are analysed with quantitative calculations.

Five samples used in this work, labelled by A, B, C, D, and E, were grown by a solid-source molecular beam epitaxy (MBE) system with the Stranski–Kranstanov growth mode. Samples A and B were both grown at 600°C with 22 periods of Ge and Si bilayers, and the Ge coverages of the two samples were 15 and 6 Å, respectively. For sample B, it is only with Ge strain layer. Samples C, D, and E all were grown at 540°C with 10 periods of Ge and Si bilayers, and the Ge coverages of these samples were 12, 15 and 18 Å, respectively. The Si layer thickness of 20 nm was used for all superlattices. Figure 1 shows a typical atomic force microscopy (AFM) image of the self-assembled Ge dots grown at 540°C, the average dot height and base are determined to be 10 and 90 nm, respectively. Raman scattering measurement was performed using a JY T64000 Raman system at room temperature. All the spectra were excited by the 488 nm line of an Ar ion laser in the backscattering, the spectral resolution

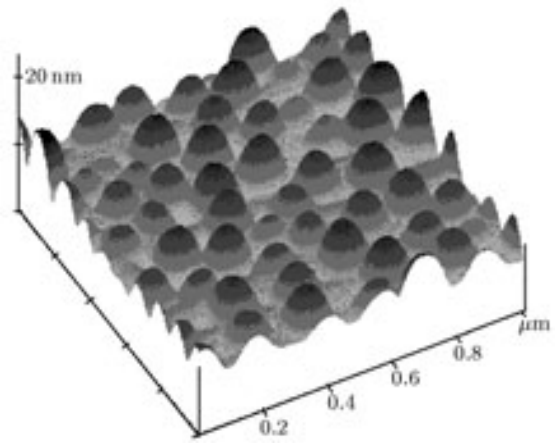

Fig. 1. A typical atomic force microscopy image of the self-assembled Ge quantum dots.

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is about $0.5\,\mathrm{cm}^{-1}$.

Figure 2 shows the Raman spectra of samples A and B. In the curve of sample A, besides the strong Si substrate signal at $520\,\mathrm{cm}^{-1}$, the Ge-Ge, Si-Ge, and local Si-Si (Si-$\mathrm{Si_{LOC}}$) vibrational peaks can be seen at 299, 417, and $436\,\mathrm{cm}^{-1}$, respectively. The appearance of the Si-Ge and Si-$\mathrm{Si_{LOC}}$ vibrational peaks implies the formation of SiGe alloy in the wetting layers and the existence of strain in Si underneath the dots. The Ge-Ge peak arises from the Ge dots, which denotes the optical mode of the dots. For sample B, the Ge-Ge mode is much weaker. The difference of the Ge-Ge modes between the two samples suggests that the Ge-Ge mode is mainly from the Ge dots rather than the wetting layer.

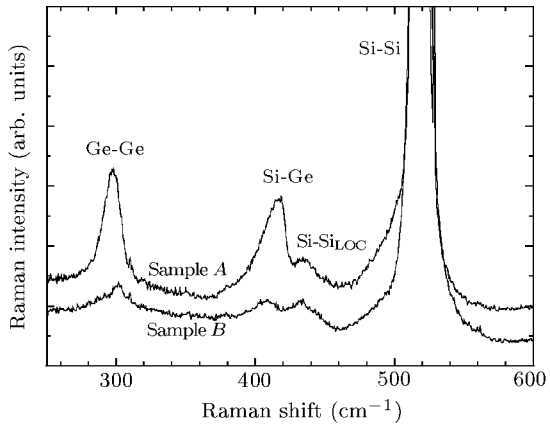

Fig. 2. Raman spectra of the samples A and B. Samples A includes 22 periods of Ge dots, while sample B just with Ge strain layers.

Figure 3 shows the Raman spectra of samples C, D, and E in the spectral region of Si-Ge and Ge-Ge optical phonons. Similar Si-Ge and Ge-Ge lines are observed for these samples. The vertical dotted line is fixed at $\omega_0 = 300\,\mathrm{cm}^{-1}$, which represents the optical phonon position for bulk crystalline Ge. The frequency positions of the Ge-Ge optical phonons in dot samples are at about $300\,\mathrm{cm}^{-1}$, the exactly values for samples C, D, and E are 299, 298, and $297\,\mathrm{cm}^{-1}$, respectively.

There are mainly two possible physical origins, in principle, which can cause a Raman shift of optical phonons. The first one is resulted from the strain: the lattice mismatch of Si and Ge leads to a compressive strain on the dots in the lateral directions, which induces a Ge-Ge mode shift to the higher frequency side. The second reason is the phonon confinement from the spatial limitations of superlattices, causing a shift of optical phonons towards lower frequency.

The blueshift of Ge optical mode due to the strain within the Ge quantum dot superlattices can be expressed as[6]

$$\Delta\omega_{\mathrm{strain}} = \frac{1}{\omega_0}\Big(q - \frac{C_{12}}{C_{11}}p\Big)\varepsilon_{xx}, \qquad (1)$$

where ω_0 is the frequency of the Ge zone-centre LO phonon; p and q are the Ge deformation potentials; C_{11} and C_{12} are the elastic coefficients; ε_{xx} is the biaxial strain. Here $\omega_0 = 0.565 \times 10^{14}\,\mathrm{s}^{-1}$, $p = -4.7 \times 10^{27}\,\mathrm{s}^{-2}$, $q = -6.167 \times 10^{27}\,\mathrm{s}^{-2}$, $C_{11} = 1288\,\mathrm{kbar}$, $C_{12} = 482.5\,\mathrm{kbar}$. For fully strained pure Ge on Si, $\varepsilon_{xx} = (a_{\mathrm{Si}} - a_{\mathrm{Ge}})/a_{\mathrm{Si}} = -0.042$ with a_{Si} and a_{Ge} being the lattice constants of Si and Ge, $\Delta\omega_{\mathrm{strain}} = 17.4\,\mathrm{cm}^{-1}$ is obtained. However, as a matter of fact, the dots in the samples are not fully strained due to the strain relaxation from the atomic intermixing at the Si/Ge interface. Thus ε_{xx} should be written as $(a_{\mathrm{Si}_{1-x}\mathrm{Ge}_x} - a_{\mathrm{Ge}})/a_{\mathrm{Si}_{1-x}\mathrm{Ge}_x}$, where $a_{\mathrm{Si}_{1-x}\mathrm{Ge}_x}$ is the lattice parameter of the SiGe alloy with the Ge concentration x and can be determined by Vegard's law $a_{\mathrm{Si}_{1-x}\mathrm{Ge}_x} = xa_{\mathrm{Ge}} + (1-x)a_{\mathrm{Si}}$. The Ge concentration x can be calculated from the relative intensity of Ge-Ge and Si-Ge optical modes peak in Raman spectra. For samples C, D, and E, $x = 0.5$ and thus $\Delta\omega_{\mathrm{strain}} = 8\,\mathrm{cm}^{-1}$.

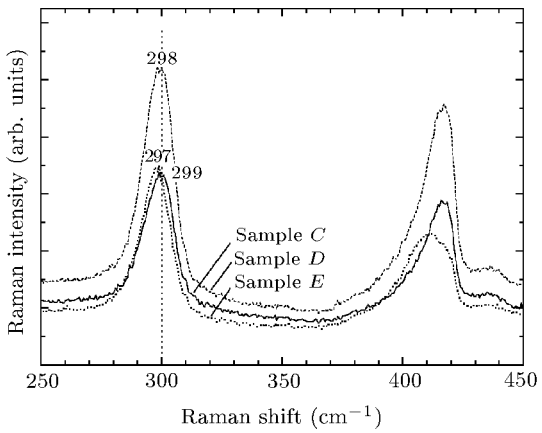

Fig. 3. Raman spectra of the samples C, D, and E. The frequency positions of the Ge–Ge peak in the samples are shifted slightly to their bulk value ($300\,\mathrm{cm}^{-1}$, the vertical dotted line).

In the present experiment, the Ge optical mode was found at about $300\,\mathrm{cm}^{-1}$ in the Raman spectra. It is the phonon confinement that causes a redshift of Ge optical mode. From the simple linear chain model, it can be easily understood why the redshift come into being: the spatial limitations of superlattices (the height of the dots) cause a shift of confined optical phonons towards lower frequency. From the Richter–Wang–Ley model (RWLM),[7,2–4] which was once successfully applied to study the phonon confinement in Si, the frequency redshift due to phonon confinement in Ge QDs approximately can be estimated. Using the RWLM, the frequency redshift can

be expressed as

$$\Delta\omega_{\text{ph-con}} = -A\left(\frac{a_{\text{Ge}}}{d}\right)^{\gamma}, \qquad (2)$$

where d is the height of the Ge dots, a_{Ge} is the Ge lattice constant, and A and γ are the constant parameters with $A = 52.3\,\text{cm}^{-1}$ and $\gamma = 1.586$, respectively. Employing the average dot height 10 nm into Eq. (2), we obtain the redshift of the samples equal about $1\,\text{cm}^{-1}$. Compared with the blueshift caused by strain, this value is a minor one, i.e., the calculated value of Ge optical phonon frequency is still larger than the experimental one. Therefore, it is suggested that there are some additional strain relaxation mechanisms besides Ge/Si interdiffusion, such as strain relaxation from dot morphology transition. For example, the more the Ge QDs transform from the pyramid shape to the dome shape, the more the strain relaxation exists.

In summary, we have reported on the investigation of Raman scattering in the self-assembled Ge quantum dot superlattices, the Si-Si, Ge-Ge, Si-Ge and Si-Si_{LOC} peaks were found in the Raman spectra, which arise from the Si substrate, the optical phonon mode of Ge, the SiGe alloy in the wetting layers and the existence of strain in Si underneath the dots, respectively. The effects of the phonon confinement and the strain within the Ge dots can induce the redshift and blued-shift of the optical phonon modes. Strain relaxation in Ge QD superlattices is not only from Ge/Si interdiffusion but also from other reasons such as dot morphology transition.

References

[1] Wang K L, Liu J L and Jin G 2002 *J. Cryst. Growth* **237** 1892
Venezuela P et al 1999 *Nature* **397** 678
Schmidt O G et al 2000 *Appl. Phys. Lett.* **77** 4341
[2] Zi J et al 1996 *Appl. Phys. Lett.* **69** 200
[3] Zi J, Zhang K and Xie X 1997 *Phys. Rev.* B **55** 9263
[4] Paillard V et al 1999 *J. Appl. Phys.* **86** 1921
[5] Liu J L et al 1999 *Appl. Phys. Lett.* **74** 1863
[6] Liu J L et al 2002 *J. Appl. Phys.* **92** 6804
[7] Richter H, Wang Z P and Ley L 1981 *Solid State Commun.* **39** 625