Semiconductor Physics, Quantum Electronics & Optoelectronics. 2003. V. 6, N 2. P. 164-168.

PACS: 63.22.+m, 68.37.Ps, 68.65.Hb, 72.10.Di

Investigation of the optical and acoustical phonon modes in $Si_{1-x}Ge_x$ QD SLs

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Abstract. Single- and multilayer structures with $Si_{1-x}Ge_x$ nanoislands have been investigated using the Raman scattering technique. The values of the mechanical strain and composition were determined in the islands of the both structures. For multilayer structure a low-frequency Raman spectrum was obtained due to the scattering on folded acoustical phonons. The experimental values of the peaks are compared with those derived theoretically.

Keywords: QD, Raman scattering, AFM.

Paper received 12.02.03; accepted for publication 16.06.03.

1. Introduction

Most of heterostructures demonstrating novel electronic and optoelectronic functions, especially those based on quantum-size effects, are realised on III-V group elements. However, modern semiconductor electronics is based on silicon with its highly developed, reliable and relatively cheap fabrication techniques. Therefore, for further increase in integration density, operation frequency and efficiency of light emission, novel concepts are required in Si-electronics. Quantum dot (QD) heterostructures are an example of such a concept. Due to their atom-like energy spectrum, these structures are promising candidates for future electronics and optoelectronics [1-3].

Quantum dots (or islands) are formed by MBE-method in the Stranski-Krastanow mode, which provides a transition from 2D layer-by-layer to 3D growth due to the reduction of the strain energy of the system. Although a lot of work has been done on the investigation of self-assembling and self-organisation of the islands, physical principles of their nucleation and growth are not completely understood yet. Thus, there is no exact quantitative description of the influence of strain on an enhanced diffusion of substrate atoms into the islands.

In this work, Raman scattering (RS) technique and atomic force microscopy (AFM) were employed for investigation of six-fold stacked structure with $Si_{1-x}Ge_x$ QDs embedded in Si matrix. A comparative analysis has been performed with the results on the analogues single layer structure.

2. Experimental technique

The structures under investigation were obtained by molecular beam epitaxy on Si(001) substrate with 250 nm thick silicon buffer layer. The islands were formed of 7.5 monolayers (ML) of Ge deposited on the buffer layer at 600 °C. After Ge deposition was finished, the islands were covered with 26 nm of silicon, to form the so-called spacer layer. The procedure was repeated 6 times. Analogous single layer structure was grown for comparison. The samples used for AFM measurement had an uncovered top island layer. Raman measurements were performed in quasi-backscattering geometry. Optical phonon modes were recorded using T64000 Jobin Yvon triple spectrometer equipped with CCD camera for multichannel detection. The low-frequency acoustical bands were taken with double DFS-24 monochromator with photomultiplier and photon counting system. In the latter experiment the sample was kept in vacuum in order to avoid air related Raman peaks in the low-frequency range. Ar^+-laser with 488 nm line was used for excitation. AFM images were taken with Nanoscope III-a operating in a tapping mode.

3. Results and discussion

A strong vertical self-ordering was found to take place in the multilayer structures with QDs. When the spacer thickness is small enough, the islands in the upper layer nucleate and grow exactly above those in the lower layer (Fig. 1). The reason is that the stretched parts of the spacer lattice above the islands are the most preferable places for accumulation of Ge adatoms because of fitting lattice parameter [3-5].

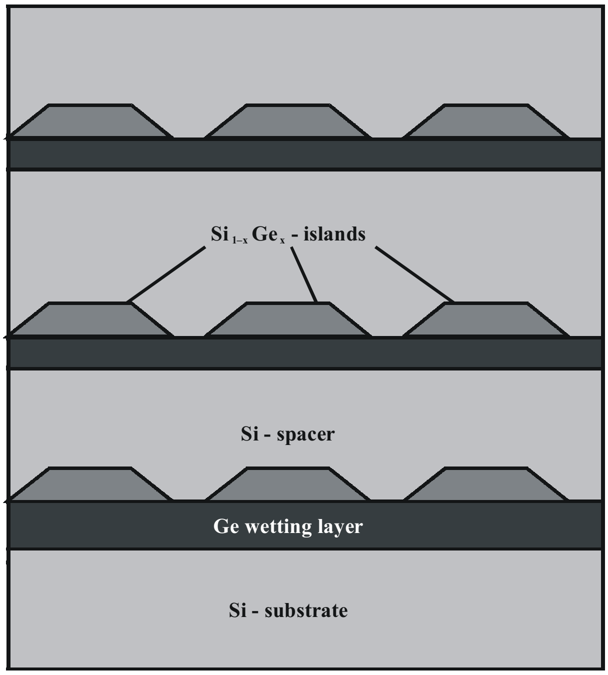

Fig. 1. A schematic cross-section of the stack with $Si_{1-x}Ge_x$ nanoislands.

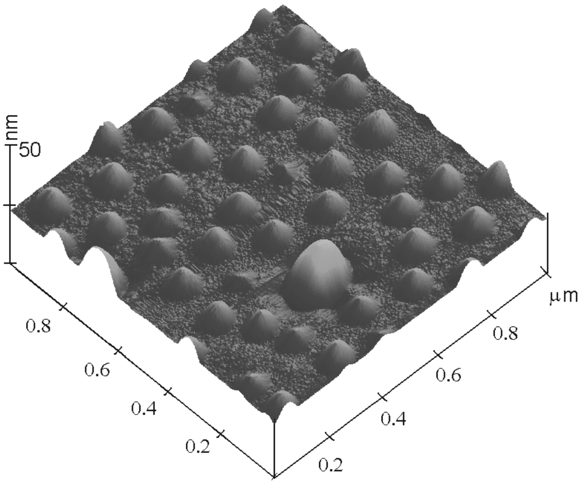

Fig. 2. AFM-image of the top island layer of the stack with $Si_{1-x}Ge_x$ nanoislands.

It was shown for a single layer structure with $Si_{1-x}Ge_x$ islands [6-9], that both composition $\boldsymbol{x}$ and strain $\boldsymbol{e}$ in the islands can be successfully determined from the positions of Raman peaks corresponding to Ge-Ge, Ge-Si and Si-Si vibration modes. We have neglected the effect of confinement of optical phonons on the position and shape of Raman peaks, because of sufficiently large average lateral dimension and height of QDs. Those were determined from AFM images (Fig. 2) and were equal 50–70 nm and 5–6 nm, respectively. The substrate contribution into the Raman spectrum of the single layer structure can be always substracted. In the multilayer structure, all spacer layers contribute to the Raman spectrum, and the problem with subtraction is, therefore, somewhat more complicated. A predominantly tensile strain in the spacers causes a low-frequency shift of the silicon spacer optical mode from that of the substrate. But the value of this shift does not exceed few cm^{-1} and can be neglected, because it is small compared to the composition-induced shift of this mode within the islands. Therefore, when x is not very close to 0 or 1, the values of the Si-Si mode can be reliably used for calculation of x and ε. It should be noted that the calculation with only Ge-Ge and Ge-Si modes leads to larger errors, because these modes have similar dependencies on composition and strain as opposed to Si-Si mode that has an inverse dependence. The frequency position of the Ge-Si mode is known to be linearly dependent on the composition only for $0 \le x \le 0.4$ [10]. We have extrapolated this dependence for an unstrained alloy by a quadratic polynomial for the whole compositional range $0.4 \le x \le 1$. After a complementary linear strain dependence of the band is taken into account, the resultant formula is:

$$\omega_{GeSi} = 387 + 81x - 78x^2 - 575\varepsilon.$$

The equations for Ge-Ge and Si-Si modes were taken from [6]:

$$\omega_{GeGe} = 282.5 + 16x - 385\varepsilon,$$

$$\omega_{SiSi} = 520.5 - 62x - 815\varepsilon.$$

Substituting experimentally found values $\omega_{GeGe} = 298.8\ cm^{-1}$, $\omega_{GeSi} = 416\ cm^{-1}$, $\omega_{SiSi} = 496.5\ cm^{-1}$ (Fig. 3, curve 1) and solving graphically the system of equations, as shown in [6], we obtain $x = 0.6$, $\varepsilon = -0.015$. The calculation by the ratio of integrated intensities of bands corresponding to the Ge-Ge and Ge-Si modes by the formula

$$I_{Ge\text{-}Ge}/I_{Ge\text{-}Si} = x/2(1-x)$$

gives the close value $x = 0.65$.

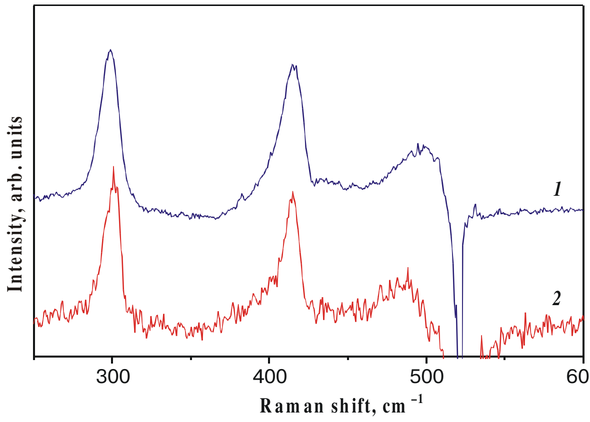

Fig. 3. Raman spectra of the stack (*1*) of the single layer structure with $Si_{1-x}Ge_x$ nanoislands (*2*). The spectrum of the silicon substrate was substracted in both cases.

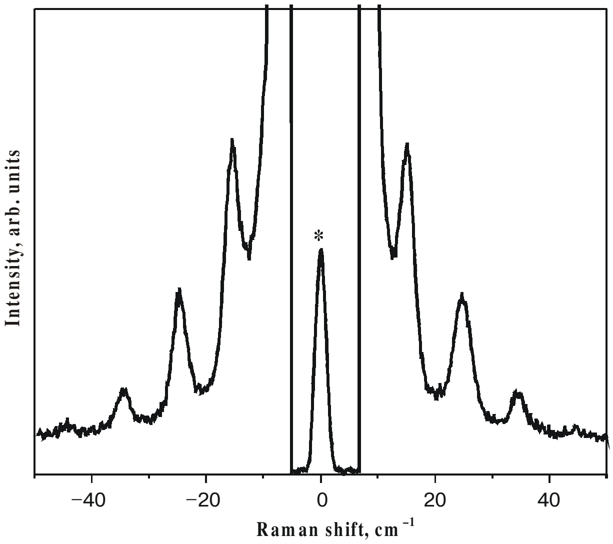

Fig. 4. Stokes and antistokes Raman spectra from the scattering on the folded LA phonons in the six-fold stack with $Si_{1-x}Ge_x$ nanoislands.

The scattering from Ge-wetting layer (WL) is forbidden in the 001(100, 100)$00\bar{1}$ geometry according to selection rules [11]. An interface mode that may manifests itself when an abrupt interface exists, in our case decreases in intensity, because of smearing the interface between Si spacer and Ge WL due to Si-Ge interdiffusion during growth at 600 °C. Even if this mode exists, its frequency is ~400 cm^{-1} [12], but in our spectrum $\omega_{GeSi} = 416$ cm^{-1}. Therefore the interface mode can be neglected in our case as well.

In order to determine the influence of the silicon spacer layers on the island parameters, we studied a single layer structure with uncaped $Si_{1-x}Ge_x$ islands. Raman spectrum of the structure is shown in Fig. 3, curve 2. The composition and strain were obtained similarly to the stack structure and were equal: $x = 0.75$, $\varepsilon = -0.017$. A higher value of Si content in the islands in the stack is due to an additional Si atom in-diffusion during the spacer growth. As a result, these islands are expected to be more relaxed compared to uncovered ones. The observed close values of a residual strain in both the capped and uncapped islands can be caused by the compensating compressive strain induced by silicon spacer on the islands in the stack.

In the low-frequency region of the Raman spectrum, a series of distinct equidistant bands is observed (Fig. 4). Such features are known to result from the scattering on folded longitudinal acoustical (FLA) phonons in SLs [1, 13, 14]. Nevertheless, Rytov's model [15] was designed for infinite and planar multilayers and was successfully applied to III-V [4] and Ge/Si [16] multilayers of QDs, containing only 5–10 QD layers. Cazayous et al. proposed the mechanism for the Raman scattering in QD multilayers, which accounts for the RS in such structures whatever is the number of layers [17-19]. The deformation-potential electron-phonon interaction was considered with taking into account the fact that due to the three-dimensional electronic confinement, translation invariance is lost and Raman scattering by all acoustic phonons becomes allowed. Therefore, all features in the low-frequency RS spectra can be interpreted as a result of RS interference that occurs when acoustic phonons interact with an ensemble of localised electronic states. The RS interference model gives not only peak frequencies but intensities including ratios and fine structures as well. At the same time it was shown in [19] that this model and more simple Rytov's model give the same peak frequencies for layer number Ni (5÷6).

In this reason the dispersion curves of our QD SL we calculated by Rytov's theory. They are depicted in Fig. 5. An artificial periodicity in SL in the growth direction produces a folding of an initial acoustical phonon dispersion curve into a minizone with $q_{max} = \pi/d$. The propagation of the phonons in the multilayer structure may be

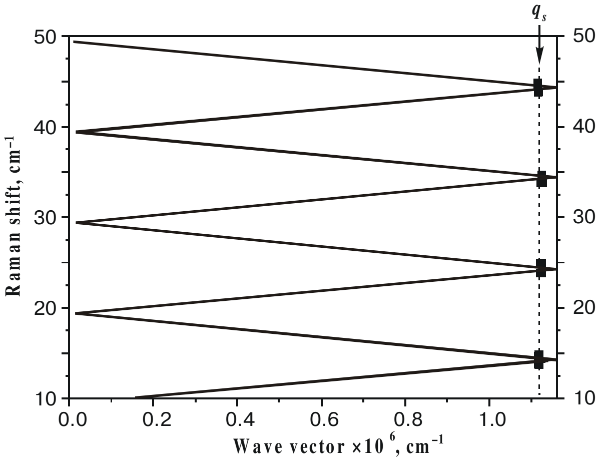

Fig. 5. The theoretical dependence of the folded LA phonon frequency on the wave vector for the six-fold stack with $Si_{1-x}Ge_x$ nanoislands.

Table 1. The theoretical and experimental Raman frequencies of the folded LA phonons in the six-fold stack with $Si_{1-x}Ge_x$ nanoislands.

	Raman frequency, cm^{-1}							
m	–1	+1	–2	+2	–3	+3	–4	+4
Calculated	14.7	15.1	24.7	24.9	34.6	35	44.6	45
Experimental	15.1	24.8	34.6	44.7				

described successfully by Rytov's theory [15] based on the elastic continuum model:

$$\cos(qd) = \cos\left(\frac{\omega d_1}{V_1}\right)\cos\left(\frac{\omega d_2}{V_2}\right) - \\ -\frac{R^2+1}{2R}\sin\left(\frac{\omega d_1}{V_1}\right)\sin\left(\frac{\omega d_2}{V_2}\right),$$

where $R = \frac{V_1\rho_1}{V_2\rho_2}$, V_1, V_2 – sound velosity; ρ_1, ρ_2 – density; d_1, d_2 – thickness of the layers 1 and 2, respectively; $d = d_1 + d_2$ – period of SL; ω – frequency of the propagating phonon; q – its wave vector. With squares in Fig. 5 marked are doublets that manifest themselves in the Raman spectrum at a wave vector of laser light $q_s = 4\pi n_\lambda/\lambda =$ $= 1.14\cdot10^6$ cm^{-1}, where λ is the laser wavelength, n_λ – the average value of refractive index for the whole structure (n_l = 4.33 for λ = 488 nm). In our case, when elastic properties of the layers are close, a reductive formula can be used: [20]:

$$\omega = V_{SL}\left(\frac{2\pi}{d}\right)m \pm V_{SL}q_s,$$

where $V_{SL} = d\left(\frac{d_1^2}{V_1^2} + \frac{d_2^2}{V_2^2} + \left(R + \frac{1}{R}\right)\frac{d_1\,d_2}{V_1V_2}\right)^{-1/2}$

Raman spectra have been modelled, as a superposition of two groups of LA phonons, propagating in the growth direction separately through the islands and WL, respectively. The values of a sound velocity for $Si_{1-x}Ge_x$-alloy islands were interpolated between those of Si and Ge. The theoretically obtained phonon frequencies are in a good agreement with those obtained experimentally (Table 1).

4. Conclusions

Using Raman scattering technique, we have shown that the silicon content in the $Si_{1-x}Ge_x$ islands of the multilayer structures is higher than that for the structure with the single layer of islands. Nevertheless, the residual strain has almost the same value in both cases. The increase of the Si content in stacked islands could enable their better relaxation, but the absence of a free surface in these islands makes their additional relaxation impossible. The narrow enough widths of the folded LA phonon bands in the Raman spectrum are an evidence of a good quality of the structure. The calculated values of these peaks are in a good agreement with the experimental ones.

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