Temperature dependence of the quantum dot lateral size in the Ge/Si(100) system

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The temperature dependence of morphological characteristics of Ge/Si(100) quantum dots grown by molecular beam epitaxy with constant growth rate at different substrate temperatures is investigated. The mean lateral size of quantum dots at 6.2 ML is shown to increase at rising substrate temperature. Comparison of experimental and theoretical results obtained in the frame of kinetic model is performed.

Modification of electronic properties of material in quantum dot (QD) heterostructures offers exciting opportunities for their use in novel optoelectronic devices [1]. Molecular beam epitaxy (MBE) and related growth technologies for the direct QD formation utilize the effect of a spontaneous islanding in strained heteroepitaxial systems. Carrier confinement in QD leads to the dependence of the wavelength of QD-based light emitting devices on the lateral size of islands. A technological need for tuning optical properties of QD arrays stimulates the studies of growth conditions influence on the surface morphology [2]. In this work we report the results of theoretical and experimental studies of the substrate temperature dependence of the QD lateral size in the Ge/Si(100) system.

The kinetic model of a stress-driven QD formation in Stranski–Krastanow growth mode [3] provides the following description of the process. The surface is stable against islanding as long as wetting layer thickness $h$ is lower than the equilibrium thickness found from the Müller–Kern criterion [4]

$$h_{eq} = \frac{k_0 \ln \left( \frac{\Phi}{h_0(1 - z(\theta)) \lambda \epsilon_0^2} \right)}{2}.$$  

Here $k_0$ is the coefficient of relaxation of deposit–substrate attractive forces, $\Phi$ the wetting energy, $h_0$ the monolayer (ML) height, $z(\theta)$ the relative relaxation of elastic energy in the island depending on contact angle $\theta$ [5], $\lambda$ the elastic modulus of the deposit and $\epsilon_0$ the lattice misfit. The onset of 2D–3D transformation occurs at critical WL thickness $h_c = (1 + \zeta^{*})h_{eq}$ at time $t^* = \zeta^{*}t_{eq}$, where $t_{eq} = h_{eq}/V$ is the time to grow the equilibrium WL, $V$ the growth rate in ML/s and $\zeta^* = 0.24A^{1/2}/B\ln^{1/2}Q$. Constants $A = \sigma(1/\cos \theta - 1)/\lambda(1-z(\theta))$ and $B = (h_{eq}/k_0)(1 - z(\theta)) \lambda \epsilon_0^2 k_0^2/2k_0T$ are determined by the ratio of surface energy to thermal energy and of elastic energy to thermal energy, respectively, $\sigma$ being the surface energy of deposit, $l_0$ the 2D lattice spacing, $T$ the substrate temperature and $\sigma = (6h_0 \cot \theta l_0^{1/3})$ a geometrical factor. If the stress-driven diffusion flux of material from wetting layer to islands
constitutes the dominant QD growth mechanism [3, 6], the control parameter is \( Q = t_0^2/\tau \) where \( \tau = 3k_0^2/8\pi b_0^2vD(T) \), \( D(T) \) is the coefficient of volume diffusion in the wetting layer and \( v \) the cutoff parameter for elastic stress field. The temperature dependence of the diffusion coefficient is approximated as \( D(T) = D_0\exp(-T_0/T) \) with \( T_0 \) being the characteristic diffusion temperature. All presented expressions are written for pyramid-shaped islands with square base of length \( L \) and aspect ratio \( \beta = \tan\theta/2 \). WL thickness is measured in the numbers of ML. As follows from definitions for time scale hierarchy of different stages of QD formation process as described in the text are presented in Table 2. Results of AFM observations and theoretical calculations for the temperature dependence of the mean lateral size of islands are shown in Fig. 2. After the end of nucleation stage, the island surface density remains constant, WL thickness decreases and the mean lateral size of islands grows with time. The evolution of mean lateral size is given by

\[
\frac{t-t'}{t_k} = \ln\left[\frac{(1+l+l^2)^{1/2}}{1-l}\right] - \sqrt{3}\arctan\left(\frac{2l+1}{\sqrt{3}}\right) + \frac{\pi}{2\sqrt{3}}, \quad l' \leq t \leq 3t_k.
\]

Here \( l(t) = L(t)/L_R \), \( l(t) \) is the mean lateral size in nm and \( L_R \) is the lateral size in the end of relaxation stage defined as

\[
L_R = 0.2a_0^2 B^{1/2} Q^{1/2} \Theta^{1/2}.
\]

The characteristic time of the size relaxation stage \( t_k = (0.47/\ln^{1/3}(Q))\beta \). The presented results are valid under the condition \( F = (5/2)\ln Q \gg 1 \) providing the applicability of classical nucleation theory and the time scale hierarchy of different stages of QD formation process \( \Delta t \ll t_k \ll t' \). The large parameter of the theory \( t' \) is the activation barrier for island nucleation at critical WL thickness. In all presented expressions, the time moment \( t = 0 \) corresponds to the equilibrium wetting layer thickness \( h_0^\text{eq} \) the overall deposition time thus being \( t + t_0^\text{eq} \). After the end of the relaxation stage (\( t > 3t_0^\text{eq} \)), islands continue to grow as \( L(t) = L_0(\nu/t^*)^{1/3} \) until the growth is interrupted.

Four samples of Ge islands on Si(100) surface were grown using a Riber SIVA (France) MBE machine at different substrate temperatures \( T = 420 \, ^\circ\text{C} \) (1), 450 \, ^\circ\text{C} \) (2), 470 \, ^\circ\text{C} \) (3) and 500 \, ^\circ\text{C} \) (4). The growth technique and characterization procedures are described in Ref. [7]. Growth rate of Ge was fixed to 0.0345 ML/s for all samples. The moment of growth interruption for each sample was related to 0.9 nm (6.2 ML) of deposited material. Corresponding deposition time is 180 s. Samples were consequently studied by atomic force microscopy (AFM) using a Digital Instruments Inc. (USA) setup.

A typical AFM image of the surface is presented in Fig. 1. It has been found that the AFM image of Ge/Si(100) nanoislands grown at 420 \, ^\circ\text{C} has a pyramid shape with a square base. At higher substrate temperatures, QD arrays contain square and rectangle base islands. Rectangle islands are elongated in \( y \)-direction (\( y \)-axis directed along [010] and \( x \)-axis along [001]), the \( L_y/L_x \) ratio ranges from 1.74 to 2.64. For all samples, the fraction of square base islands remains predominant. The aspect ratio of square base and elongated QD increases with the rise of substrate temperature. For square base islands, the variation in the aspect ratio ranges from \( \beta = 0.08 \) at \( T = 420 \, ^\circ\text{C} \) to \( \beta = 0.24 \) at \( T = 500 \, ^\circ\text{C} \). The lateral size of islands at 6.2 ML considerably increases with rising substrate temperature. The obtained experimental results are summarized in Table 1. Accuracy of measuring the island size is estimated as 5%.

Calculations were made for square base islands with the contact angle \( \theta \) selected to adjust the measured aspect ratio. The results of calculations at the parameters of the Ge/Si(100) system [6] \( (2.6 \times 10^{12} \, \text{dyn/cm}^2; \, \epsilon_0 = 0.042, \, h_0 = 0.145 \, \text{nm}, \, b_0 = 0.395 \, \text{nm}, \, \Phi = 450 \, \text{erg/cm}^2, \, \sigma = 800 \, \text{erg/cm}^2, \, k_0 = 0.9, \, \nu = 10) \) and \( T_0 = 7750 \, \text{K}, \, D(T = 470 \, ^\circ\text{C}) = 2.6 \times 10^{-13} \, \text{cm}^2/\text{s} \) for the main characteristics of island formation process described in the text are presented in Table 2. Results of AFM observations and theoretical calculations for the temperature dependence of the mean lateral size of islands are shown in Fig. 2.

To sum up, the presented theoretical and experimental results demonstrate a strong temperature dependence of morphological characteristics of Ge/Si(100) QD grown with constant growth rate at different substrate temperatures. The mean lateral size of QD at 6.2 ML is shown to increase at rising substrate...
Table 1 Results of AFM observations of QD arrays grown at different temperatures.

<table>
<thead>
<tr>
<th>sample No.</th>
<th>$T$ ($^\circ$C)</th>
<th>height (nm)</th>
<th>lateral size (nm)</th>
</tr>
</thead>
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<tr>
<td></td>
<td></td>
<td></td>
<td>square base</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>$L_x$</td>
</tr>
<tr>
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<td>4</td>
<td>500</td>
<td>5.0</td>
<td>20.2</td>
</tr>
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</table>

Table 2 Theoretical characteristics of the QD formation process at different temperatures.

<table>
<thead>
<tr>
<th>$T$ ($^\circ$C)</th>
<th>$h_{eq}$ (ML)</th>
<th>$h_c$ (ML)</th>
<th>$t_{eq}$ (s)</th>
<th>$t^*$ (s)</th>
<th>$\Delta t$ (s)</th>
<th>$t_R$ (s)</th>
<th>$F$</th>
<th>$L_R$ (nm)</th>
<th>$L(6.2 \text{ ML})$ (nm)</th>
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<td>108.4</td>
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<td>51.5</td>
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<td>10.6</td>
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<td>16.2</td>
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<tr>
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<td>5.6</td>
<td>92.0</td>
<td>71.8</td>
<td>4.4</td>
<td>16.1</td>
<td>23</td>
<td>28.0</td>
<td>20.6</td>
</tr>
</tbody>
</table>

Fig. 1 AFM image of Ge/Si(100) QD at 6.2 ML, grown at $T = 450 ^\circ$C. Scan area is 300 x 300 nm$^2$.

Fig. 2 Theoretical and experimental temperature dependences of Ge/Si(100) QD lateral size.

temperature. The results obtained might give a way to control the morphology of QD arrays which could be used for tuning the wavelength of Si-based light emitting devices.

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References