Estimation of critical thickness of Stranski–Krastanow transition in GeSi/Sn/Si system

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Abstract. In this paper Stranski–Krastanow growth of Ge$_x$Si$_{1-x}$ epitaxial layers on the Si(001) surface with pre-deposited tin layer with the thickness less than 1 ML is considered. For the calculations of critical thickness of transition from 2D to 3D growth in this paper a theoretical model based on general nucleation theory is used. This model is specified by taking into account dependencies of elastic modulus, lattices mismatch and surface energy of side facet on the composition $x$, as well as change in the adatoms diffusion coefficient and surface energy of the substrate in the presence of tin. As a result, dependencies of critical thickness of Stranski–Krastanow transition on composition $x$ and temperature are obtained. The simulated results are in a good agreement with experimentally observed results.

1. Introduction
Since 1990s Si$_{1-x}$Ge$_x$/Si material system with nano-dimensional inclusions of germanium has acquired a reputation of very promising system for various applications in electronics and optoelectronics. Great attention to the Si$_{1-x}$Ge$_x$/Sn/Si and Ge$_{1-y}$Sn$_y$/Si systems was also attracted recently because they allow one to effectively manage the band-gap energy and to obtain direct band semiconductors. These materials may be used for creation of photodetectors, solar cells and light-emitting devices and compete with traditional materials based on AIII-BV compounds [1, 2].

One of the most promising methods of creation of Si$_{1-x}$Ge$_x$/Si heterostructures is molecular beam epitaxy. During the growth of quantum dots in this method one can manage the morphology of islands’ ensemble by changing growth temperature, deposition rate, and volume of deposited material.

One of the most important values, characterizing the process of molecular beam epitaxy is critical thickness for Stranski–Krastanow transition from 2D to 3D growth. But to date there is no theoretical models capable to adequately describe initial stages of growth in GeSi/Sn/Si system and to predict values of critical thickness of 2D-3D transition and its dependencies on growth temperature and composition of solid solution.

It is well known that thin tin layer act as a surfactant during deposition of germanium on silicon [3–6]. Meanwhile, tin segregates to the surface of both silicon and germanium and increases the mobility of adatoms on the surface of substrate. Therefore, in case of GeSi layers deposition on the silicon surface with pre-deposited 0.5–1.0 ML thick tin layer it necessary to additionally take into account the increase in diffusion coefficient of atoms of growing layer on the surface of substrate covered by tin. Besides that, the presence of surfactant on the substrate’s surface alters its surface energy. In the case of tin layer on the silicon surface it is known that this energy slightly decreases [3, 4]. So, in our estimations we should also take this fact into account by changing the value of surface energy.
2. Theoretical model, results and discussion

For description of the Ge$_{1-x}$Si$_{x}$ layers deposition on the silicon surface it is essential to take into account the presence of thermodynamic parameters on the composition $x$. For this purpose the values of physical constants for pure materials (Si, Ge) and the Vegard’s law are used:

\[ \varepsilon(x) = \varepsilon_0 x, \]

\[ \lambda(x) = x\lambda(Ge) + (1-x)\lambda(Si), \]

\[ \gamma(x) = x\gamma(Ge) + (1-x)\gamma(Si), \]

where $\varepsilon(x)$ is the lattice mismatch of Ge$_{1-x}$Si$_x$ layer and silicon substrate, $\varepsilon_0$ is the lattice mismatch in the case of pure Ge, $\lambda(x)$ is the elastic modulus, $\gamma(x)$ is the specific surface energy [10].

If the modelling of quantum dots formation kinetics in Ge$_{1-x}$Si$_x$/Sn/Si system is required for estimation of surface density and size distribution function of quantum dots, it is carried out similarly to Ge/Si system [7–10] but with the parameters recalculated by the formulae described above.

For calculations of the characteristics of quantum dots growth in Ge$_{1-x}$Si$_x$/Sn/Si and Ge$_{1-x}$Sn$_x$/Si systems for small tin contents we used the following values for the parameters of the model [8–10]: $l_0 = 0.395$ nm, $d_0$ = 0.145 nm, $\gamma(Ge)$ = 800 erg/cm$^2$, $\gamma(Si)$ = 1400 erg/cm$^2$, $\lambda(Ge)$ = 1.27·10$^{12}$ dyn/cm$^2$, $\lambda(Si)$ = 1.8·10$^{12}$ dyn/cm$^2$, $\nu_0 = 0.042$, $\Psi_0 = 450$ erg/cm$^2$, $\varphi = 20^\circ$, $\nu = 10$, $D(0) = 10^{-4}\exp(-1.21/k_BT)$ cm$^2$/s.

For estimation of critical thickness of transition in the case of Ge$_{1-x}$Si$_x$ layers deposition on silicon surface with pre-deposited tin layer with the thickness of 0.5–1.0 ML it is necessary to additionally take into account the increase in diffusion coefficient of atoms of growing layer on the surface of substrate with tin. This question is poorly studied in literature but it was shown [11] that the diffusion coefficient of Ge on Si in the presence of 1 ML thick Sn layer increases by four orders of magnitude. So, for the calculations of critical thickness of transition in Ge$_{1-x}$Si$_x$/Sn/Si system we used two values of the adatoms diffusion coefficient: $D = D_010^2$ and $D = D_010^4$, where $D_0$ is the diffusion coefficient of atoms on pure Si surface. It is known that presence of Sn reduces surface energy insignificantly, but numerical values of this reduction are not given in the literature. We will assume in our calculations that surface energy of silicon decreases by 100 erg/cm$^2$ and 200 erg/cm$^2$.

It is known, that in case of Sn-mediated epitaxy of Ge on Si with the ~0.5–1 ML thick tin layer critical thickness of Stranski–Krastanow transition decreases from ~5 ML, typical for clean Si surface, to ~4 ML in the presence of tin [3–6]. Calculations with the formulae described in this article give for the critical thickness values of $h_s = 4.3$ ML and $h_s = 4.0$ ML for two stated above diffusion coefficients correspondingly, that is in very good agreement with the experimental data.

Dependencies of critical thickness of transition $x$ in Si$_{1-x}$Ge$_x$/Sn/Si system for the mentioned diffusion coefficients and surface energies at $T = 300, 500, 700$ °C were calculated (Figure 1). It is obvious, that the curve behaviors qualitatively resemble the case of deposition on clean silicon surface. This means that with increasing of Ge content $x$ critical thickness of transition decreases in the presence of Sn. But for any composition $x$ critical thickness of transition decreases in the presence of tin compared to the case of deposition in GeSi/Si system.

There are few data on the values of critical thickness of transition in Si$_{1-x}$Ge$_x$/Sn/Si system for various compositions. Figure 2 shows calculated and experimental [3] values of critical thickness of transition to 3D growth in Si$_{1-x}$Ge$_x$/Sn/Si system for different diffusion coefficients and substrate’s surface energies. Comparison with experimental results (Figure 2) indicates that theoretical predictions are in rather good agreement with experiments and describes the observed behavior of critical thickness with variation in composition correctly, especially with the assumption of the surface energy reducing by 200 erg/cm$^2$. Some discrepancy in obtained values may be caused by variations in growth rate and temperature, as well as difference in techniques of critical thickness estimation. So, it is quite probable that energetic parameters of the system slightly alter in the presence of tin and the critical
thickness increases significantly, but this question demands further experimental and theoretical investigations.

![Figure 1](https://example.com/fig1.png)  
**Figure 1.** Critical thickness of transition to 3D growth in the presence of tin with assumption of diffusion coefficient increasing by four orders of magnitude.

![Figure 2](https://example.com/fig2.png)  
**Figure 2.** Comparison of calculated critical thicknesses with experimental data [3] with assumption of diffusion coefficient increasing by two (2) and four (4) orders of magnitude and with assumption of the substrate surface energy decreasing by 100 erg/cm² (3) and 200 erg/cm² (4).

3. Conclusions
   
   Thus, in this paper theoretical model for calculation of dependencies of critical thickness of Straniski–Krstananow transition on composition $x$ for Si$_{1-x}$Ge$_x$/Sn/Si system at various temperatures is proposed. Moreover, the described model allows one to estimate surface density and size distribution function of quantum dots in these systems. Although there are a very limited number of experimental data on the critical thickness of transition from 2D to 3D growth in Si$_{1-x}$Ge$_x$/Sn/Si systems, the results of calculations demonstrate good agreement with the data available. In addition, derived expressions explain the experimentally observed temperature dependencies of critical thickness at various germanium contents.

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