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Effect of species-dependent surface atomic mobility on pattern selection during alloy growth

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Spontaneous formation of nanometer-scale composition-modulated structures is a common phenomenon for III–V and II–VI semiconductor alloys [1]. These structures are mostly formed in open systems during the epitaxial growth. It is typical for semiconductor epitaxial growth that bulk diffusivity of atoms is negligibly small compared to the surface diffusivity. For an alloy growth, composition modulation in every atomic layer is being formed at the surface during the growth of this layer and persists when this layer is buried by the subsequent layers. Modulations of composition from the entire thickness of the epitaxial film affect, via long-range strain field, the surface migration of atoms [2, 3, 4].

For alloys grown on a (001)-substrate, most of observed composition-modulated structures are 2D ones modulated in both [100] and [010] directions. The possible directions of modulation can be explained by the linear stability analysis of the homogeneous alloy growth. If the elastic anisotropy is the dominant effect compared to the surface diffusion anisotropy [4], the instability occurs for the first time in elastically soft directions [100] and [010]. Then, however, the linear stability analysis does not address the question of the selection between 1D and 2D structures. To describe the final modulated structure, a non-linear theory is needed.

In our earlier papers [S] we considered the effect of non-linear coupling between compositional and morphological instabilities on steady-state structures formed during epitaxial growth. It was shown that at sufficiently low growth velocities 2D steady-state structures form, modulated in both [100] and [010] directions. This effect is similar to that for strained islands in lattice-mismatched systems where 2D structures such as an array of pyramids provide more efficient elastic relaxation than 1D structures such as an array of prisms [S]. However, for lattice-matched systems this effect is important only for sufficiently large periods of the resulting structures, d > 700 Å. This is beyond typical scales where modulated structures could serve as quantum dots.

Among other non-linear effects which may lead to the formation of 2D structures the effect of species-dependent atomic mobility [7] is the most plausible. The Arrhenius-type behavior of the diffusion coefficient, $D(T) = D_0 \exp(-E_a/T)$, together with the dependence of the activation energy on composition $E_a = E_a(c)$ results in very strong dependence of diffusion coefficient on composition. In the present paper we consider the effect of this dependence on steady-state structures. This effect is purely kinetic and has no analogs in thermodynamics.

We consider the growth of an alloy $A_{1-c}B_cC$ by molecular beam epitaxy on an atomically rough surface. The alloy is *lattice-matched on average* to the (001)-substrate. The growth proceeds via deposition of atoms on the surface, surface migration of atoms in a stress- and composition-dependent chemical potential, and incorporation of atoms into the growing crystal, desorption being neglected. Let the composition be equal to $c(\mathbf{r}) = \overline{c} + \phi(\mathbf{r})$, the average composition being $\overline{c} = 1/2$, and, for simplicity, the surface profile be taken to be flat h(x, y) = vt, where v is the average growth velocity controlled



Fig. 1. Steady-state diagram containing the regions of homogeneous growth, of the growth of a 1D structure, and of the growth of a 2D structure.

by the deposition flux. The kinetic equation describes the evolution of composition fluctuations $\phi(\mathbf{r})$ at the advancing surface

$$\frac{\partial \phi}{\partial t} = \nabla_i \left[\frac{D(\phi)}{T} \nabla_i \frac{\delta F}{\delta \phi} \right] - \frac{v}{a} \phi \,. \tag{1}$$

Here $F = F_{\text{chem}} + F_{\text{grad}} + F_{\text{elast}}$ is the total Helmholtz free energy, where $F_{\text{chem}} \sim \int f_{\text{chem}}(c)dV$ is a chemical free energy of the alloy depending locally on the alloy composition, F_{grad} is a gradient energy, $\sim \kappa \int (\nabla \phi)^2 dV$, and F_{elast} is the elastic energy induced by composition fluctuations. The composition-dependent diffusion coefficient $D(\phi)$ is related to the substitutional diffusion of alloy components on the surface which is taken into account up to the second order in ϕ , $D(\phi) = D_0 + D_1\phi + D_2\phi^2$ and a is the lattice parameter.

The linear stability analysis [3, 5] showed that, for a given growth velocity v, the instability occurs in a certain temperature interval. The positive formation enthalpy of the alloy and the elastic interaction energy between the adatoms and the "buried" composition modulation in the entire thickness of the epitaxial film are the two driving forces of the decomposition. At high temperatures, the entropy contribution to the alloy free energy hinders the driving forces to decomposition and stabilizes the growth of a homogeneous alloy. At low temperatures, the surface diffusion is "freezed" out and does not provide formation of a modulated structure. The solid line in Fig. 1 depicts the boundary of the instability region found in [5]. The homogeneous growth is unstable at $v < v_c(T) = [4D_0(T_c - T)]/(a^2\kappa)$, where T_c is the critical temperature in the slow deposition limit [2].

In the present paper we seek the steady-state solution to kinetic equation (1) in the weak segregation regime close to the boundary of the instability region, i. e. at $v/v_c = 1 - \eta^2$, where $\eta \ll 1$. In this regime there is only one unstable mode for each elastically soft direction, and composition fluctuations can be written in the form $\phi(x, y) = 2\eta \left[(T_c - T)/a^3 \right]^{1/2} \left[\psi_x \cos(k_0 x) + \psi_y \cos(k_0 y) \right] +$ stable modes, where $k_0 = \sqrt{2(T_c - T)/(a^3 \kappa)}$. The reaction of stable modes, such as those with $\mathbf{k} = (2k_0, 0)$ and $\mathbf{k} = (k_0, k_0)$, on unstable modes can be taken into account in the adiabatic approximation.

To illustrate this reaction, we consider a composition fluctuation with the wave vector $\mathbf{k} = (k_0, 0)$ (Fig. 2(a)). This fluctuation is unstable and its amplification implies the surface flux of atoms B from A-rich domains to B-rich domains, Fig. 2(b). Assume that A-rich domains have larger atomic mobility than B-rich domains, $D(\phi < 0) > D(\phi > 0)$,



Fig. 2. Stabilizing effect due to the dependence of surface atomic mobility on composition in 1D system. (a): Unstable composition fluctuation. (b): Surface flux forming composition fluctuations of Fig. 2(a). (c): Surface atomic mobility. (d): Surface flux of Fig. 2(b) in case of varied atomic mobility (c). (e): Second harmonic of composition fluctuations due to modulated surface flux (d). (f): Surface flux induced by the second harmonic (e). (g): The first harmonic of the surface flux in case of varied atomic mobility (c).

Fig. 2(c). In this case the flux of atoms B from A-rich domains towards domain boundaries is larger than the flux from domain boundaries to B-rich domains Fig. 2(d). This results in the accumulation of atoms B at domain boundaries, i.e. in formation of a composition fluctuation with the wave vector $\mathbf{k} = (2k_0, 0)$, Fig. 2(e). Such a fluctuation is energetically not favorable and decays via surface flux out of domain boundaries, Fig. 2(f). Due to composition dependence of atomic mobility this flux is also deformed in a way that most of atoms B move towards A-rich domains, Fig. 2(g). As a result, there appears non-linear stabilizing effect. This is the action of the composition fluctuation along x-direction on itself by means of the second harmonic. There is a similar action of the composition fluctuation in the x- direction on fluctuations in the y-direction and vise versa due to the appearance of composition fluctuations with wave vectors like $\mathbf{k} = (k_0, k_0)$.

The resulting kinetic equations in the weak segregation regime have the following form, very similar to that of Ref. [5]:

$$\frac{\partial \psi_x}{\partial \tau} = \eta^2 \left[\psi_x - a_L \psi_x^3 - a_T \psi_x \psi_y^2 \right] + O(\eta^4)$$
$$\frac{\partial \psi_y}{\partial \tau} = \eta^2 \left[\psi_y - a_L \psi_y^3 - a_T \psi_y \psi_x^2 \right] + O(\eta^4) , \qquad (2)$$

where

$$a_{L} = 3 - \frac{D_{2}}{D_{0}} \times \frac{(T_{c} - T)}{A_{4}a^{3}} + \frac{D_{1}^{2}}{D_{0}^{2}} \times \frac{10(T_{c} - T)}{9A_{4}a^{3}}$$

$$a_{T} = 6 - \frac{D_{2}}{D_{0}} \times \frac{2(T_{c} - T)}{A_{4}a^{3}} + \frac{D_{1}^{2}}{D_{0}^{2}} \times \frac{2}{A_{4}a^{3}} \times \frac{(T_{c} - T)\varepsilon_{0}^{2}Ra^{3}}{\varepsilon_{0}^{2}Ra^{3} + (T_{c} - T)}$$
(3)

Here ε_0 is the lattice mismatch between pure binary constituents of the alloy, AC and BC, R is a certain combination of elastic moduli, for GaAs $R = 0.2 \text{eV}/\text{Å}^3$, and A_4 is related to the fourth-order contribution to the chemical energy, $(A_4 = 6 (\partial^4 f_{\text{chem}}/\partial^4 c) \text{ at } c = 1/2)$. If $a_L < a_T$, the steady state structure is 1D and, if $a_L > a_T$, the steady state structure is 2D. It follows from Eqs. (3) that at temperatures close to T_c the final structure is one-dimensional and at lower temperatures $(T_c - T) > \varepsilon_0^2 Ra^3$, the final structure is 2D.

This transition can be explained as follows. The species dependent atomic mobility results in "effective barriers" hindering surface atoms from migration across domain boundaries. These barriers are unavoidable for a surface flux in case of 1D structure (Fig. 2), while in case of 2D structure there is a possibility for a surface flux to go around these barriers, e.g. along < 110 >-directions. Therefore, the effect of species-dependent atomic mobility favors 2D structures. The dependence of atomic mobility on composition enhances as the growth temperature decreases. As a result, below a certain temperature the steady-state structure is 2D.

This effect is in a way complementary to the effect of surface corrugation. While the effect of surface corrugation is important in case of a large mismatch ε_0 between two binary constituents of the alloy and at a large spatial scale, the effect of species-dependent atomic mobility is stronger for moderate ε_0 and is essential at small scales. E.g. for $\varepsilon_0 = 0.08$ the final structure is 2D at periods 50 Å and smaller, and for $\varepsilon_0 = 0.02$ the final structure is 2D at d < 200 Å.

To conclude, the steady state phase diagram of Fig. 1 applies to any alloy. We show a possibility to form 2D structures with arbitrary small wavelength and explain commonly observed 2D structures modulated in [100] and [010] directions (see, e. g. [8]). Kinetic phase transition between the growth of 1D structure and the growth of 2D structure provides a possibility to tune the structure between 1D and 2D ones at arbitrary scale by varying temperature and/or growth velocity.

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