

A binary phase field crystal study for phase segregation of liquid phase heteroepitaxial growth



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ABSTRACT

The binary phase field crystal (PFC) model is employed to investigate influences of atomic sizes and mobility differences on the liquid phase heteroepitaxial. It was found that large size atoms are driven toward regions of tensile stress which correspond to peaks in a compressively strained film but to valleys in a film with tensile strain. Small size atoms are on the contrary in contrast for large size atoms. Due to the existence of vertical separation and lateral separation resulting from atomic size differences, the epitaxial layer exhibits double-island phenomenon: light and dark islands. In the presence of different mobilities, atoms with greater mobility accumulate at the film surface. And in the process of epitaxial growth, there are misfit dislocations nucleating in valleys where the strain is highest.

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1. Introduction

Due to unique optical and electronic properties, with potential applications in optoelectronics and semiconductor devices, self-assembled of nanostructure in semiconductor heteroepitaxy has been intensively studied [1–4]. These structures include junctions, quantum dot islands and superlattices in which planar interfaces are highly desired [5–7]. However, the difference of lattice constant between the deposited material and substrate results in mismatch strain. Morphological instability induced by the strain leads to nonplanar surfaces and defects in the film. This stress-driven morphological instability [8,9] has been studied extensively, and it is well established that the morphological instability can change the growth mode of a thin film from a layer-by-layer mechanism to a three-dimensional island mechanism. However, previous works mainly focus on the study of pure materials [10–12], with the development of alloy thin films, more and more investigations about alloy heteroepitaxy have been reported. A latest report on alloy heteroepitaxy from Elder [13] et al has shown influences of substrate thickness on the growth of the single island and multiple islands. Compared with pure materials, alloy thin films have more complicated compositions. As for epitaxial growth of alloy thin films, alloy components may be prone to phase separation and according to Vegard's law, the lattice constant of the film is generally a function of alloy compositions. Therefore, the growth of alloy thin films needs further study. Recently, alloy phase

separation [14–16] has captured many scholars' attentions. The main issues motivated us are that how atomic sizes and mobility differences affect processes of phase separation and further influence the stability of epitaxial thin films.

During the actual production process of thin films, it is very difficult to control the thickness of thin films because of the fast growth rate. In order to get nanoscale films, we find another way known as phase field crystal model to predict the growth of thin films. Liquid phase epitaxy as a mature technology has been used in the production of semiconductor optoelectronic devices, as well as magnetic garnets, superconductors, ferroelectrics and other optical materials. Therefore, the purpose of this paper is to illustrate how the two-dimensional binary PFC model [17,18] addresses compositional effects in alloy liquid phase heteroepitaxy, focusing on influences of atom size differences and mobility differences on phase separation.

2. Simulation method

2.1. Model

The binary phase field crystal model proposed by Elder [19] recently has not only incorporated elasticity and plasticity on atomic length and diffusive timescales and multiple crystal orientations which are caused by the periodic oscillations of the crystal lattice, but also involves the solidification, phase segregation, solute diffusion and other important features. For a binary alloy made up of A and B atoms, to define the total number density $\rho = \rho_A + \rho_B$. To simplify calculations it is convenient to fix

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introduce the following dimensionless fields: $n_A = (\rho_A - \bar{\rho})/\bar{\rho}$, $n_B = (\rho_B - \bar{\rho}_B)/\bar{\rho}$. Also, it indicates that the atomic number density field n and the concentration field φ (To show the atomic density difference, it can be approximated as the concentration field), which gives

$$\begin{aligned} n &= n_A + n_B = (\rho - \bar{\rho})/\bar{\rho} \\ \varphi &= (n_B - n_A) + (\bar{\rho}_B - \bar{\rho}_A) = (\rho_B - \rho_A)/\bar{\rho} \end{aligned} \quad (1)$$

where F is the free energy functional given by

$$F = \int d\bar{x} \left(\frac{n}{2} \wedge n - \frac{t}{3} n^3 + \frac{v}{4} n^4 + \gamma \varphi + \frac{w}{2} \varphi^2 + \frac{u}{4} \varphi^4 + \frac{K}{2} |\bar{\nabla} \varphi|^2 \right) \quad (2)$$

where $\wedge \equiv B_0^l + B_2^l \varphi^2 + B_0^s (2\nabla^2 + \nabla^4) + 4\alpha \varphi B_0^s (\nabla^2 + \nabla^4)$. Briefly outlining the physical meaning of the model parameters is as follows: the $B_0^l, B_2^l, B_0^s, t, v, w, K, \gamma, \alpha$ and u are constants, whose values depend on the material parameters. t, γ and v are model parameters associated with the Taylor coefficient. The length scale of phase separation is decided by the interplay of u, K and w . B_2^l determines liquid volume modulus with the changing of atomic concentration field. B_0^l is the liquid temperature. B_0^s is associated with the solid phase of the elastic constants. $\Delta B = B_0^l - B_0^s$ corresponds to temperature expressing the initial driving force of crystallization, which can be enhanced by lowering ΔB . α represents the solute diffusion factor measuring the size difference between A and B atoms, and is defined as $\alpha = (a_B - a_A)/a_0$ where a_A, a_B , and a_0 are the relaxed-state lattice parameters for pure A , pure B and the alloy film. Also, in the spirit of keeping calculations as simple as possible without losing the basic physics contained in the model, $\gamma = 0$ in the free energy. Minimizing the free energy, $n = A \left[1/2 \cos(2qy/\sqrt{3}) - \cos(qx) \cos(qy/\sqrt{3}) \right]$ will be used. Substituting this expression into (2) and minimizing with respect to q and A , recalling that φ is assumed constant over the scale that n varies, getting $q_{eq} = \sqrt{3}/(2R)$ and $A_{min} = 4(t + \sqrt{t^2 - 15v\Delta B})/(15v)$.

To simulate microstructure formation in binary alloys, dynamical equations of motion for the fields n and φ need to be developed.

$$\begin{aligned} \frac{\partial n}{\partial t} &= \nabla \left(M_1 \nabla \frac{\delta F}{\delta n} \right) + \nabla \left(M_2 \nabla \frac{\delta F}{\delta \varphi} \right) = \frac{\partial n_A}{\partial t} + \frac{\partial n_B}{\partial t} = M_A \nabla^2 \frac{\delta F}{\delta n_A} + M_B \nabla^2 \frac{\delta F}{\delta n_B} \\ &= (M_A + M_B) \nabla^2 (\wedge n - tn + vn^3 + \gamma) + (M_B - M_A) \nabla^2 K \nabla^2 \varphi \\ &\quad + (M_A - M_B) \nabla^2 (n^2 B_2^l \varphi + 2\alpha n^2 B_0^s (\nabla^2 + \nabla^4) + w\varphi + \mu\varphi^3) \end{aligned} \quad (3)$$

$$\begin{aligned} \frac{\partial \varphi}{\partial t} &= \nabla \left(M_2 \nabla \frac{\delta F}{\delta n} \right) + \nabla \left(M_1 \nabla \frac{\delta F}{\delta \varphi} \right) = \frac{\partial n_A}{\partial t} - \frac{\partial n_B}{\partial t} = M_A \nabla^2 \frac{\delta F}{\delta n_A} - M_B \nabla^2 \frac{\delta F}{\delta n_B} \\ &= (M_A - M_B) \nabla^2 (\wedge n - tn + vn^3 + \gamma) - (M_B + M_A) \nabla^2 K \nabla^2 \varphi \\ &\quad + (M_A + M_B) \nabla^2 (n^2 B_2^l \varphi + 2\alpha n^2 B_0^s (\nabla^2 + \nabla^4) + w\varphi + \mu\varphi^3) \end{aligned} \quad (4)$$

where $M_1 = (M_A + M_B)/\rho^2$ and $M_2 = (M_B - M_A)/\rho^2$. To get Eqs. (3) and (4), we used the approximate value of Eq. (1): $n = n_A + n_B$, $\varphi = n_A - n_B$. Eqs. (3) and (4) couple the dynamics of the fields n and φ through a symmetric mobility tensor. The dependence of mobility M_A and M_B in general depends on local crystal density and the local relative concentration of species A and B . While M_A and M_B are different, a simple Euler algorithm was used for the time derivative and the spherical Laplacian approximation was used.

Assuming a substitutional diffusion between species A and B , that the same M mobility applies for the two species, furthermore, considering that the mobility coefficient is a constant, the

dynamics of the fields n and φ decouple, the respective equations of motions have the form:

$$\frac{\partial n}{\partial t} = M \nabla^2 \frac{\delta F}{\delta n} \quad (5)$$

$$\frac{\partial \varphi}{\partial t} = M \nabla^2 \frac{\delta F}{\delta \varphi} \quad (6)$$

The numerical discretization of Eqs. (5) and (6) is implemented using the mixed explicit-implicit Fourier transformations and operator splitting by Tegze et al. [20].

2.2. The chosen of parameters

The equilibrium diagram is shown in Fig. 1 for the appropriate parameters set as follows: $B_0^s = 1.0$, $B_2^l = -1.8$, $t = 0.6$, $L = 2.65$, $\mu = 4.0$, $\alpha = 0.26$ and $w = 0.088$. Our simulation is conducted on a periodic system of size $512\Delta x \times 512\Delta x$, the binary alloy film with average density difference $\varphi = 0$ grows from a liquid phase above the bulk coherent spinodal temperature, the time step $\Delta t = 0.5$. According to the equilibrium diagram, we choose $\Delta B_0 = 0.00886$ to implement all of the following simulations, growth at this ΔB_0 above the miscibility gap is typical of experimental conditions and should ensure that phase separation is driven by local stresses and is not due to spinodal decomposition. Initial conditions consisted of a binary unstrained crystalline planar substrate of eight atomic layer thickness in the bottom of the simulation area which is below a symmetric supercooled liquid of components A and B . In what follows the misfit strain is defined as $(a_{film} - a_{sub})/a_{sub}$, for a symmetric mixture of A and B components $a_{film} = (a_A + a_B)/2$.

3. Results and discussion

For the reason that the evident contrast as bright and dark regions can highlight the phase separation in the alloy, grey-scale maps are adopted. The results are presented in Fig. 2 for two components with different atomic sizes and same mobility. Fig. 2 (a) and (c) show the atomic number density difference while Fig. 2(b) and (d) demonstrate the local concentration difference. As shown in Fig. 2(a) and (b), at the initial stage of growth, epitaxial

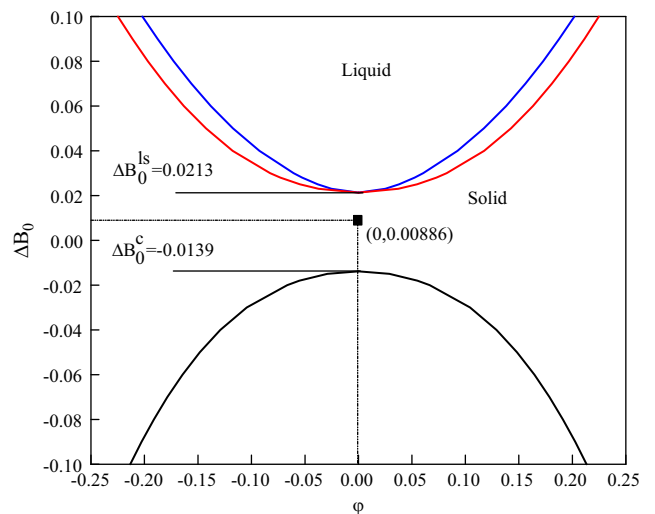


Fig. 1. Phase diagram as a function of $\Delta B_0 - \varphi$ for two-dimensional triangular system. The blue solid line corresponds to liquid phase, the red solid line corresponds to solid phase, the black square dot corresponds to selected initial condition. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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