A Novel Way to Fabricate Nanowires by Directed Self-Organization of Atoms

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We propose here a novel way to fabricate self-assembled nanowire structures on atomically flat substrates during epitaxial deposition. A phase field model is developed for the free energy of the system, which includes short-range as well as long-range elastic interactions between deposited atom clusters and mediated by the substrate. We show here that a weak external periodic field can be utilized to guide the formation of nano-wires on atomically smooth substrates by the self-assembly of deposited atoms. In particular, we show that a weak sub-surface strain field generated by buried interfacial dislocations can dramatically control self-organization of deposited atoms and the emerging atom cluster length scale. The atom composition and field requirement conditions for nucleation of sharp nano-wires are shown in this study.

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I. INTRODUCTION

The potential utilization of self-organization phenomena in epitaxial thin film growth has now become an important direction in nano-science research to manufacture ultra small structures for future electronic and photonic devices. Nanowires have been the object of intense theoretical and experimental investigations because of their unusual properties in thermoelectronic devices[1, 2].

Since traditional lithographic methods to fabricate nanowires have size limitations [3], more research has recently been directed to self-assembly approaches, such as chemical synthesis [4], vapor-liquid-solid growth techniques [5] and atom deposition methods [6]. However, realistic advances in this technology much depends on providing the means for atoms to assemble themselves in a precise manner, and to control their size distribution during the fabrication process. The fabrication of massive numbers of well-aligned nanowire structures on 2-D substrates is still clearly challenging, and requires both theoretical and experimental advances.

The assembly on individually deposited atoms into self-organized structures on a substrate results in atomic lusters with characteristic length scale that is typically two orders of magnitude larger than atomic dimensions. Atomic cluster shapes and length scales are generally imprecise for nanotechnology. Thus, there is a need to control both the shape and length scale of atomic clusters. One solution is to impose an external field that provide constraints for precise self-assembly of atomic clusters. Recently a possible solution to the regularized nanowire fabrication proposed by Liu[7] and Venezuela[8] is to generate a step-patterned surface during step flow on a vicinal surface. The nanopattern of surface steps is formed by relatively uniform step bunching arrays resulting from interlayer step-step elastic interactions. Since step bunches are much straighter than normal surface steps, it can be an efficient constraint on the nucleation of adatoms.

In addition to surface patterning, external fields provide promising candidates to regulate the self-assembly of nanostructures. Guiding fields can be imposed either directly on adatoms (such as electromagnetic fields [9–11]) or mediated through the substrate (such as elastic fields by interfacial dislocations [12, 13]). In the latter case, experimental observations indicate that the applied external field that exhibits a significant effect on self-organization is very weak. For example, in the experiments conducted by Kim et. al [12?], the external field is induced by misfit interfacial dislocation arrays. The strain field of buried interfacial dislocations is estimated to be on the order of $\nu b/h$, where ν is Poission's ratio, b is Burger's vector and h is the thickness of the substrate layer on the surface. In recent experiments on Ge self-assembled quantum dots on partially relaxed SiGe buffer layers [12, 13], the buffer layer thickness is about $80\,\mathrm{nm}$, and the length of the Burger's vector is about $0.2\,\mathrm{nm}$. Thus, the strain magnitude is only on the order of 0.1%. Thus, the diffusion energy barrier for an unclustered single atom changes only by $0.01\,\mathrm{eV}$ or less [14–16]. This value is obviously much smaller than strain-free diffusion barriers. In other words, although the change in the diffusion energy of adatoms is very small, the influence of a weak external field has already been experimentally demonstrated [12, 13].

We present here a continuum phase-field approach to describe the evolution of spatially organized atomic clusters as a result of interactions between deposited adatoms and between adatoms and substrates. The resulting self-organization pattern is understood as an underlying instability as a consequence of the interplay between local interfacial interactions of cell boundaries and global substrate-mediated interactions. We distinguish the effects of externally-applied elastic fields from intrinsic long-range fields generated by cluster-cluster interac-

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tions. One of the main objectives of the present work is to show that a nonuniform ultra-weak external strain field can dramatically change the self-organized pattern and length scale of atomic clusters resulting from atomic deposition.. Such external fields can be used as templates for "directed" self-organization of surface clusters. It will be shown that if the conditions are selected carefully, the imposed weak strain field can be utilized to fabricate well-aligned nanowire structures.

In section II, a formulation that includes long-range (L-R) substrate-mediated interactions and the presence of a nonuniform external elastic field is developed. We then discuss in section III the nature of the external-field generated from a buried interfacial dislocation network in Si-Ge heterepitaxial strutures, and show how nanowire structures can be formed by "directed" self-organizations. Conclusions are finally given in section IV.

II. FORMULATION

Surface atomic clusters are formed on substrates by the process of physical vapor deposition, where single atoms in the vapor phase impinge on a substrate, stick and diffuse on the surface, leading to the nucleation and growth of atomic clusters. When the process is performed at low pressures and with thermalized atoms (i.e. without high energy neutrals or ions), the substrate surface structure is not changed, and atomic diffusion is dominated by hops on the surface, without the assistance of near surface point defects. We assume here that the substrate surface is atomically smooth, without reconstruction or surface steps. When adatoms are deposited on the surface, surface diffusion of adatoms is the dominant transport process and thus interdiffusion of adatoms through vapor and through the substrate will be ignored. A continuous model for the concentration of surface adatoms can be constructed within the framework of chemical kinetics via the following mass conservation equation:

$$\frac{\partial c}{\partial t} = \mathcal{R}(c) - \boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{J}} \tag{1}$$

where $c(\mathbf{r}, t)$ is the concentration of adatoms on the substrate. The reaction rate of adatoms is represented by $\mathcal{R}(c)$. If linear adsorption and desorption rates (α and β) are assumed, then $\mathcal{R}(c) = \alpha(1 - c) - \beta(c)$. The diffusion flux $\boldsymbol{\mathcal{J}}$ is determined by linear non-equilibrium thermodynamics. With Onsager's principle for an isothermal process, and the fact that the chemical potential is the functional derivative of the free energy, we have the following expression for the atomic mass flux $\boldsymbol{\mathcal{J}}$ for a single specie adsorbate layer[17].

$$\boldsymbol{\mathcal{J}} = -L\boldsymbol{\nabla}\mu_c = -L\boldsymbol{\nabla}\frac{\delta\mathcal{F}}{\delta c} \tag{2}$$

where L is the atomic mobility ($L = D/k_{\mathrm{B}}T$), and $D = \rho_0 D_s$ is the diffusion coefficient [18]. $\mathcal{F}$ is the free energy of adsorbed atoms, which is composed of three components:

$$\mathcal{F} = \mathcal{F}^{(a)} + \mathcal{F}^{(s)} + \mathcal{F}^{(c)} \tag{3}$$

where $\mathcal{F}^{(a)}$ is the free energy associated with direct interaction (nearest-neighbor) between adatoms, $\mathcal{F}^{(s)}$ is the free energy for their interaction with the substrate, and $\mathcal{F}^{(c)}$ is the free energy for the indirect (through substrate and long-range) interaction between clusters of atoms.

The nearest-neighbor free energy $\mathcal{F}^{(a)}$ can be formulated either by variational mean field theory [19, 20] or by Bragg-williams approach [21]. Both methods result in the same expression for $\mathcal{F}^{(a)}$ as a continuous surface coordinate ($\mathbf{r}(x, y)$) as [20]

$$\mathcal{F}^{(a)} = \int_S d\mathbf{r} \left[k_{\mathrm{B}}Tf(\mathbf{r}) - \frac{1}{2}\epsilon_0 c^2(\mathbf{r}) + \frac{1}{2}\xi_0^2|\nabla c(\mathbf{r})|^2 \right], \tag{4}$$

where $f(\mathbf{r}) = [1 - c(\mathbf{r})]\ln[1 - c(\mathbf{r})] + c(\mathbf{r})\ln[c(\mathbf{r})]$. The chemical potential of this nearest-neighbor (N-N) interaction is

$$\mu^{(a)} = k_B T \ln\left(\frac{c}{1-c}\right) - \epsilon_0 c - \xi_0^2 \nabla^2 c. \tag{5}$$

Next, let's consider the calculation of $\mathcal{F}^{(s)}$, the free energy associated with ad-atom interaction with a strained substrate. When a layer of atoms is adsorbed on a substrate at equilibrium, the discontinuity of the intrinsic surface stress between adatom clusters and the substrate provides a large driving force for self-organization [22, 23]. On the edge of a cluster, the geometric discontinuity can be replaced by a pair of tangential force dipoles[24]. The relation between the force density (f_α) and surface intrinsic stress ($\sigma_{\alpha\beta}$) is given by

$$f_\alpha(\mathbf{r}) = \frac{\partial \sigma_{\alpha\beta}(\mathbf{r})}{\partial x_\alpha}. \tag{6}$$

where α and β denotes the coordinate indices (1 or 2) on the surface. In our monolayer model, if a commensurate growth is reasonably assumed and the intrinsic stresses of clusters are treated in an effective manner, we have a first order approximation[25], expressed by Vegard's law in which the stress is assumed to be linearly dependent on concentration:

$$\sigma_{\alpha\beta}(\mathbf{r}) \approx \sigma_{\alpha\beta} c(\mathbf{r}), \tag{7}$$

where $\sigma_{\alpha\beta}$ is considered a material constant on the homogeneous substrate surface in commensurate heteroepitaxial structures. In the isotropic case, we have

$$\sigma_{\alpha\beta} = \sigma\delta_{\alpha\beta}. \qquad (8)$$

where σ denotes a scaler material constant which is about $0.1\,\text{eV}$ per Åfor Ge. The free energy induced by the substrate can be generally expressed as the force times the displacement ($\mathbf{u}$)

$$\mathscr{F}^{(s)} = -\int_S d\mathbf{r}\, f_\alpha\, u_\alpha = -\int_S d\mathbf{r}\, [\sigma\nabla_\alpha c(\mathbf{r})]\, u_\alpha \qquad (9)$$

Thus, using the interchangable property of derivatives and variational operators, and integrating by parts with Gauss's theorem, the chemical potential simply becomes

$$\mu^{(s)}(\mathbf{r}) = \sigma u_{\alpha,\alpha} = \frac{1}{2}\sigma\left[\varepsilon_{xx}(\mathbf{r}) + \varepsilon_{yy}(\mathbf{r})\right]. \qquad (10)$$

where $\varepsilon_{\alpha\beta}$ is the external strain field applied in the substrate. Since the elastic energy per adatom is approximated as $\frac{1}{2}\sigma_{\alpha\beta}\,\varepsilon_{\alpha\beta}$, it is easy to see that atomic clusters are considered as part of the substrate surface that store elastic energy. If this interaction is uniform, there is of course no effect in the adatom dynamics (Equation 1). However, when the substrate elastic field is nonuniform, the term $\mu^{(s)}$ will act as a source term in the governing equation (Equation 1). However, it should be noted that since this term appears in the gradient of a current, this does not affect the mean coverage. It also indicates that the rigorous formulation of system dynamics should include the deformation of the substrate coupled with adatoms dynamics. In the case of a weak elastic field, the "flat-surface" assumption is a good approximation.

Finally we consider the free energy due to cluster-cluster interactions. This can be formally expressed by the Green's function method with substitution of equation (7) in (6) without any external strain field on the substrate:

$$\mathscr{F}_2^{(s)} = -\frac{1}{2}\int_S\int_S d\mathbf{r}\,d\mathbf{r}'\sigma_{\alpha\mu}\nabla_\mu c(\mathbf{r})G_{\alpha\beta}(\mathbf{r}-\mathbf{r}')\sigma_{\beta\nu}\nabla'_\nu c(\mathbf{r}')$$
$$(11)$$

where the energy is defined positive for attractive interactions, and we perform a double surface integral in equation (11). $G_{\alpha\beta}(\mathbf{r}-\mathbf{r}')$ is the surface Green's function which denotes the displacement component α at position $\mathbf{r}'$ caused by a unit point force acting at position $\mathbf{r}$ in direction β. Considering the symmetry of Green's Functions and applying the same techniques used in deriving equation (10), then using the elastic isotropic condition (8), we obtain a coverage-dependent part of the chemical potential ($\mu_c^{(s)}$) as

$$\mu_c^{(s)} = \sigma^2\int_S d\mathbf{r}'\left[\nabla_\alpha G_{\alpha\beta}(\mathbf{r}-\mathbf{r}')\,\nabla'_\beta c(\mathbf{r}')\right]. \qquad (12)$$

Substituting equations (5), (10) and (12) into equation (2), and then into (1), we finally obtain a "phase field" kinetic equation for the continuum concentration c as

$$\partial_t c = \frac{1}{\tau}(c_0 - c)$$
$$+ \frac{D_0}{k_B T}\nabla^2\left[k_B T\ln\left(\frac{c}{1-c}\right) - \epsilon_0 c - \xi_0^2\nabla^2 c\right.$$
$$+ \frac{1}{2}\sigma a^2\left[\varepsilon_{xx}(\mathbf{r}) + \varepsilon_{yy}(\mathbf{r})\right]$$
$$\left.+ \sigma^2 a^4\int_S d\mathbf{r}'\,\nabla_\alpha G_{\alpha\beta}(\mathbf{r}-\mathbf{r}')\,\nabla'_\beta c(\mathbf{r}')\right] \qquad (13)$$

where $c_0 = \alpha/(\alpha+\beta)$, and $\tau^{-1} = \alpha+\beta$. The concentration, c, is in units of atom/atom. The introduction of a^2, where a is the lattice constant, is for consistency of units in Equation 13.

III. RESULTS AND DISCUSSIONS

A good candidate of external fields to guide nanowire structure formation is interfacial dislocations in heteroepitaxial structures. It is experimentally shown that a modulated surface strain field can be produced by an array of buried 60-degree misfit dislocations on a partially relaxed semiconductor buffer layer [12, 13]. For cubic semiconductors, mixed edge-screw dislocation lines lying at the interface between a thin film and (001) substrate run along $\langle 110\rangle$ directions and form rectangular arrays with spacing in the micron range [12]. According to the experiments by Kim et al.[12], the fabrication is carried out in two steps. First is the deposition of Ge atoms with a slow growth rate of $00.05\,\text{Å/s}$ at $700°C$. The second step is the quenching process where the sample is quenched to room temperature. The growth pattern is finally formed at room temperature. Thus in our model, we can reasonably ignore the details of the growth process. The deposition and desorption during the self-organization phase can be considered to have reached equilibrium. Our simulation starts from an initial random small perturbation in the spatial concentration.

The elastic field of a buried dislocation array can be formally obtained using 2-D Fourier transforms in isotropic or anisotropic single layers[26], or in multilayer systems[27]. However, we develop here a simpler solution for a single infinite straight dislocation in a homogeneous isotropic half space. Using the complex variable representation method[28], we represent the strain field with a simple equation:

$$\varepsilon_{xx} = \frac{2}{\pi}\frac{1}{(1-2\nu)}\frac{(b_x x + b_z h)xh}{(x^2+h^2)^2} \qquad (14)$$

In deriving the interaction part of the chemical potential (equation 10), we may note that the term $\mu_e^{(s)}$

is considered only when the field from the substrate is nonuniform. This means that a uniform strain field on a *flat* substrate surface, such as that due to coherent lattice mismatch in heteroepitaxial structures, will not influence self-organized patterns on the surface in our model. Thus we investigate the same material (Ge) deposited on a Ge substrate with straight parallel dislocation lines inside. The Ge lattice constant a is $0.566\,$nm at room temperature. We take the pair potential ϵ_0 as about $0.3\,$eV. The adsorption and evaporation rates, α and β, respectively, are assumed to be $1.67\times10^{-5}\,$cm^{2}s^{-1} and $1.67\times10^{-6}\,$cm^{2}s^{-1} with an approximate deposition rate of $1\,\mu m$ per minute [19]. The surface diffusion of Ge adatoms is strongly dependent on the temperature T and the coverage c. For simplicity, we assume that the surface diffusion coefficient D_s is constant and that $D_s = D_0 = 2.57\times10^{-7}$cm^{2}s^{-1}[12]. The intrinsic surface stress σ is 100 meVÅ^{-2} for Ge(001). The elastic stiffness coefficients for $\nu = 2.75\times10^{10}\,erg\cdot$cm^{-3} [29]. The edge components of the Burger's vector of an interfacial dislocation in a partially relaxed SiGe buffer layer are estimated as $b_x \approx -1.93\,$Å and $b_z \approx -2.73\,$Å, with the negative sign indicating that the extra half plane is extending away from the surface downwards to the infinity.

Because of the convolution form of the integral in equation (13), a convenient way to solve the kinetic partial differential equation is the Fourier spectral method. Let's denote the wave-vector as $\mathbf{q}$, its amplitude as q, and the concentration in the transformed domain as c_q, then by taking the Fourier Transform (FT) of equation (13), we get

$$\begin{aligned}
\partial_t c_{\mathbf{q}} = {}& \frac{1}{\tau}(c_{0\mathbf{q}} - c_{\mathbf{q}}) \\
& - \frac{D_0}{k_{\mathrm{B}}T}q^2\left[\frac{1}{2}\sigma a^2\mathrm{Tr}(\boldsymbol{\varepsilon})_{\mathbf{q}} + k_{\mathrm{B}}T\left\{\ln\left(\frac{c}{1-c}\right)\right\}_{\mathbf{q}}\right. \\
& \left. - \left(\epsilon_0 + \xi_0^2 q^2 + \sigma^2 a^4 q_\alpha q_\beta G_{\alpha\beta}(\mathbf{q})\right)c_{\mathbf{q}}\right]
\end{aligned} \qquad (15)$$

where $G_{\alpha\beta}(\mathbf{q})$ is the surface Green's function in Fourier space, which is evaluated by an analytical solution of general Green's functions in anisotropic substrates with cubic symmetry [30]. It is noted that the numerical technique relies on the application of periodic boundary conditions. We use a Galerkin approach to solve equation (15) in transformed space. Moreover, a pseudo-spectral technique is applied to nonlinear terms in the equation and a split-radix FFT package is adopted to implement the transformations [31].

First we start by presenting results of computer simulations for the surface density profile, c, in a one-dimensional system, with an initial uniform coverage of $c_0 = 0.2$, and 0.015, respectively. Results of our simulations are shown in Figure (1A) for an initial coverage of 0.2, and in Figure (1B) for an initial coverage of 0.015. The strain field generated by a straight interfacial dislocation line below the 40 nm buffer layer is illustrated
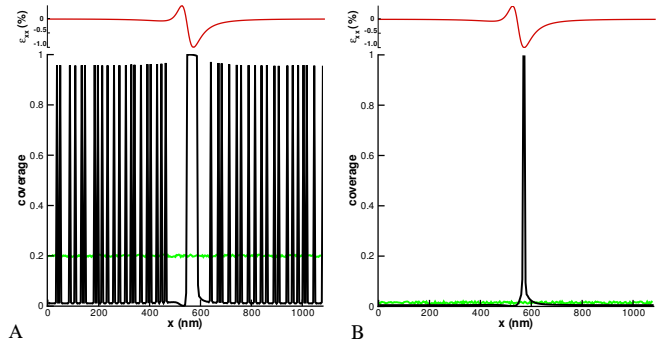

FIG. 1: Spatial distribution of coverage (fractional surface concentration of adatoms) for an initial concentration of (A) $c_0 = 0.2$, (B) $c_0 = 0.015$, where in both figures, the grey lines denote the initial profiles with random perturbations and the black line denotes the equilibrium profiles.

above of coverage profile. It can be seen from Figure (1A) that in the case of $c_0 = 0.2$, the spontaneous self-organization pattern is biased by the weak strain field with thickening of the cluster on the compressive side of the dislocation. Two denuded zones are generated on both sides of the dislocation. If c_0 is reduced below some critical level, say down to 0.015, a dramatic change occurs in which the weak strain field of the buried dislocation sweeps out all the coverage pattern, and strengthens the only mode around the compressive region, as shown in Figure (1B). It should be noted that when c_0 is reduced further, the initial random perturbation decays and the system returns to its initial stable state.

When the system is extended to two dimensions, internal self-organization cannot be represented by "peaks" as in the 1-D case. If the temperature is below the critical value as numerically calculated by [19, 32], a hexagonal dot pattern and/or a stripe pattern natually emerge [20]. The critical temperature can be obtained by linear stability analysis and is described by a marginal stability curve [19]. Within the instability region, the pattern transitions can be analyzed by weakly nonlinear stability analysis [19]. When an external field is present, the dots and stripes of atomic clusters are guided and redistributed by superimposing the external field profiles. In another words, a higher density of dots or stripes will exist in the maximum compressive strain field and a lower density in tensile strain region. This is exactly what we observe in the present simulations. As shown in Figure 2, when $c_0 = 0.5$, the system without external fields forms a nanostripe pattern. As a result of initially random perturbations, stripe orientations are randomly distributed all over the surface as long as it is elastically isotropic. Even if some preferential orientations of stripes can be formed by the elastic anisotropy of the surface [20, 33], the longitudinal length and spacing between stripes are at the nano-scale, and cannot be considered as nanowire structures. However, when a weak external field is included, the nanosized stripes are redistributed. In an equilibrium state as shown in Figure (2D), stripes are
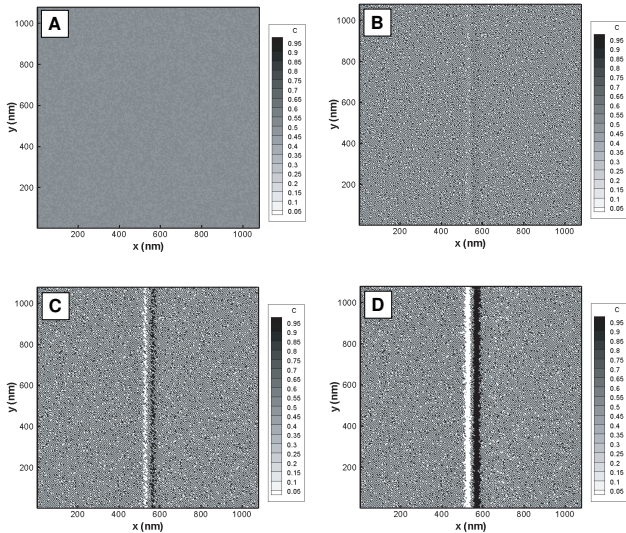
FIG. 2: 2-D simulation of directed self-organization from an initial random perturbation at $c_0 = 0.5$ with $40\,\mathrm{nm}$ buffer layer thickness, where (A), (B), (C) and (D) show the results at $0\,\mathrm{sec}$, $0.22\,\mu\mathrm{sec}$, $2.75\,\mu\mathrm{sec}$ and $13.75\,\mu\mathrm{sec}$, respectively.

FIG. 3: 2-D simulation of directed self-organization from an initial random perturbation at $c_0 = 0.05$ with $40\mathrm{nm}$ buffer layer thickness, where (A), (B), (C) and (D) show the results at $0\,\mathrm{sec}$, $0.055\,\mu\mathrm{sec}$, $0.55\,\mu\mathrm{sec}$ and $7.15\,\mu\mathrm{sec}$, respectively.

densely packed along the dislocation line. Despite a shallow region of a denuded zone, most stripes will exist in the open region of the decaying strain field. A similar situation occurs in the case of the dot-pattern regime of self-organization [20]. For example, when c_0 are around 0.1, dots which is only along the dislocation line will achieve a higher density, while dots far away from the line will decay.

However, when the uniform coverage c_0 is small enough but not so small as to result in a stable system, an interesting phenomenon similar to the 1-D case takes place. Under these restricted conditions, all atom clusters will uniformly agglomerate along the dislocation line, and small clusters that used to exist in the open regions away from the vicinity of the dislocation line are completely swept off due to the influence of the very weak dislocation field. Figure 3 shows our simulation results in the case of $c_0 = 0.05$. It can be noted that under these condition, the weak external field along one direction provides a pattern change from dots to a well-aligned stripe that runs along the dislocation line.

The interesting thing in both 1-D and 2-D simulations is that an external field exhibits a global effect over micron length scale, although the governing equation (Equation 13) is based on local balance and interactions. In spite of the weakness of the dislocation strain field, the sharp alignment phenomena observed in Figure 1B and 3 indicates that the wavevectors associated with the dynamics are locked by the characteristic wavevector of the applied strain field under certain combinations of control parameters. Similar to the concept of frequency locking of a periodic forced oscillator [34, 35], this wavevector locking or resonance in the adatom dyn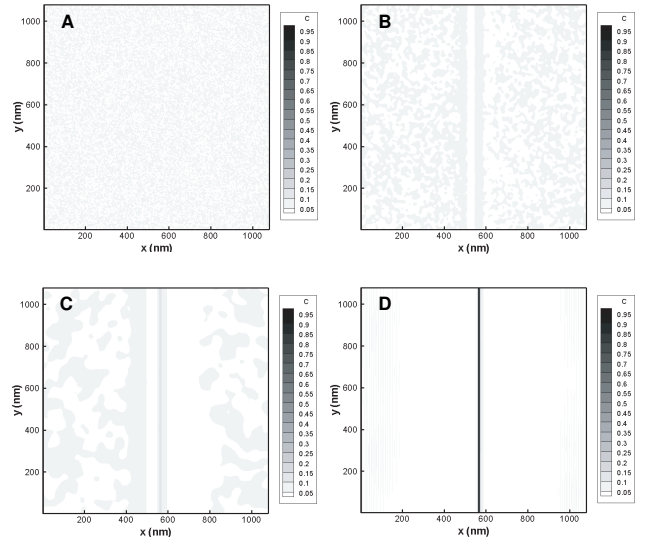amic system cannot be explained by a simple superposition, and certainly results from nonlinear effects mainly through the logarithmic and cluster-cluster interaction terms. In other words, the interactions between two different scales (nanoscale and microscale) are connected by nonlinear bifurcations.

A detailed discussion for the analysis of locking in spatially extended systems is out of the scope of the present paper. However, since this scale bridging is consistent with the character of nanowire structures in which the lateral extent of atom assembly is constrained to tens of nanometers or less, while the self-assembly process is unconstrained in one dimension, , it is proposed here that this sharp alignment can be utilized for the precise fabrication of nanowires. Because our phase field model is based on a monolayer structure of clusters, it should be understood that it is applicable during the every early nucleation stage for cluster evolution (e.g. in the Stranski-Krastanov growth regime after the wetting layer is deposited and before the multilayer island structure is formed). Since island nucleation is mainly determined by monolayer growth and that 3-D construction of islands does not have a dramatic effect on the selected patten of nanostructures or on its stability, it would be a promising way to adjust a proper combination of control parameters to realize the desired formation of nanowires if a corresponding external field network is provided.

IV. CONCLUSIONS

In this work, we developed a phase field model that describes the evolution of surface adatom atomic clusters

on atomically smooth substartes. The model shows that the self-assembly process of adatoms is in fact a competition between two length scales. The first is a "natural" length scale in the nano-meter range, and arises from local interactions and overall reduction of system energy by clustering at this particular length scale. The second scale is associated with an imposed external and interactive field, such as that supplied by a buried or interfacial dislocation. However, to utilize this idea in the precise fabrication of nano-wires requires careful control of system parameters. the most significant control parameters are the temperature and initial coverage. Above a critical temperature, the system is totally stable against small concentration fluctuations. the same is also the case for very low initial coverage. If the initial coverage is in a restricted range, and the system temperature is below the critical value, small initial perturbations result in an instability that sweeps long wave-vectors (i.e. nano-scale patterns), and re-inforces short wave-length vectors corresponding to the imposed weak field. It is proposed that this technique can have wide applications by coupling weak electromagnetic, electric, or elastic external fields to adatom deposition for fabrication of on-demand nano-structures, such as nanowires and quantum dots.

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