Domain competition during ballistic deposition: Effect of surface diffusion and surface patterning

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(Received 30 March 2011; published 4 August 2011)

We investigate domain competition occurring during aggregate growth under ballistic deposition on a one-dimensional substrate by kinetic Monte Carlo simulations. In order to capture adsorbate molecules being deposited vertically, domains grow tall by extending their branches laterally and suppress the growth of neighboring short domains. When molecules are deposited on a flat substrate and frozen at the deposition site, the population density of domains, ρ, decreases by a power law as $\rho \sim h^{-2/3}$ at height h. In contrast, if the effect of surface diffusion is taken into account, the domain density decreases rapidly as $\rho \sim 1/h$.

On a substrate patterned with an array of nanopillars, domains growing from pillar tops tend to envelop those growing from gaps between pillars. To completely suppress the growth of domains in gaps, pillar periodicity λ should be smaller than a critical value λ_c. We estimate this value approximately using the slope angle and the aspect ratio of a single isolated domain.

DOI: 10.1103/PhysRevE.84.021601 PACS number(s): 68.43.Jk, 05.70.Ln, 68.55.J−

I. INTRODUCTION

In heteroepitaxial crystal growth, a lattice mismatch between the substrate and the epitaxial layer induces several defects such as threading dislocations or cracks in overlayers. These defects deteriorate the quality of overgrown epitaxial layers. In order to reduce such defects, several approaches have been proposed, such as the fabrication of low-temperature buffer layers [1,2] and epitaxial lateral overgrowth [3]. The buffer layer is supposed to consist of thin pillars, on which further epitaxial layers are grown at a high temperature [4]: Since the overlayer is not in direct contact with the substrate, it is expected to be free from misfit strains and possess high crystalline quality. Recently, various nanoheteroepitaxial methods have been proposed for application to microelectronics. It has been reported that the nanoheteroepitaxy on dense arrays of substrate nanopillars relaxes elastic strain and reduces various defects [5]. Another study on growth on nanoporous substrates also confirmed the reduction in dislocation densities [6,7]. In these studies on nanoheteroepitaxy, the epitaxial layer grew mainly on top of the patterned substrate without filling gap spaces among pillars [5] or in nanopores [7].

Motivated by these studies, in the present work we studied domain competition during epitaxial growth on a nanopatterned substrate surface. We adopted a ballistic deposition (BD) model because the usual solid-on-solid (SOS) model for crystal growth is inappropriate in the present case since voids or overhangs are prohibited in the SOS model. In contrast, the BD model allows the formation of overhangs, which provide empty spaces under solidifying molecules.

In the BD model, molecules being deposited solidify at the highest position when they come in contact with the substrate or already solidified adsorbate [8,9]. An adsorbed molecule deposited on the substrate acts as a nucleation center and defines a domain. Then, an adsorbate overlayer is divided into several domains, each of which grows by incorporating molecules being deposited. As domains grow, they extend their branches laterally to capture many molecules incident from above; thus, domains compete with each other for molecules being deposited. When one of the domains overgrows and covers a neighboring one, the supply of molecules to the latter is cut and it ceases growing. Thus, during the growth of the adsorbate overlayer, domains coarsen and their population density decreases. In this study, we investigate this domain coarsening analytically and numerically by performing kinetic Monte Carlo (KMC) simulations.

In the case that the substrate surface is patterned with an array of pillars, domains growing from the pillar tops have an advantage over those growing from the bottom of pillar gaps: Since the former have less contact with the substrate than the latter, they are relatively free from misfit strains. Therefore, it is favorable to stop the growth of domains from the gap bottoms. For achieving this, the pillars should be high and densely populated. We determine the maximum periodicity of the pillar array for this growth termination as a function of pillar height.

In the BD model, deposited molecules are frozen at the sites of their first contact with the substrate and/or adsorbed overlayer, and thus, the formed overlayer contains numerous vacancies with a ramified growth front. The structure of the porous aggregate does not correspond to the dense structure of adsorbate crystals obtained in previous studies [5–7]. We expect that allowing lateral diffusion of isolated molecules on the adsorbed overlayer would result in the packing in the overlayer becoming dense and the growth front becoming smooth [10]. Therefore, we include lateral diffusion in the KMC simulations and study its effect on domain competition. We find that surface diffusion favors domains growing on pillar tops and enhances the maximum periodicity for suppressing gap domains.

II. BALLISTIC DEPOSITION ON A FLAT SUBSTRATE

A. Ballistic deposition without diffusion

We consider BD growth in a square lattice and measure the length in the unit of a lattice constant, $a = 1$. At the bottom of the simulation box of size $L \times H$ lies a one-dimensional substrate with a length L. Adsorbate molecules are deposited

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vertically in the $-y$ direction onto a flat substrate, and at the instant when they touch down on the substrate or adsorbed overlayer, they freeze at that site. The grown adsorbate eventually form a porous aggregate with a density less than unity, but it is compact and not fractal. The interesting feature of BD aggregation is the self-affine character of its growth front [8,9]. The growth front is characterized by a set of the highest aggregate positions $y = h(i)$ at each column $x = i$. Then, the average height is defined as $h = L^{-1}\sum_{i=1}^{L} h(i)$ and the front width is given as $W = \sqrt{L^{-1}\sum_{i=1}^{L}[h(i) - h]^2}$. When h is low, W increases as $W \sim h^{\beta}$ with the growth exponent β, whereas for large h, it saturates at $W \sim L^{\alpha}$ with the roughness exponent α. The crossover occurs at a height $h \sim L^{z}$, where $z = \alpha/\beta$ is the dynamic exponent. The height dependence of the front width is summarized by the scaling relation

$$W = L^{\alpha} f\left(\frac{h^{1/z}}{L}\right). \tag{1}$$

Here, $f(x)$ is a scaling function that is constant for large $x \gg 1$ and is proportional to x^{α} at small x [8].

We now study the domain competition during the BD. When a deposited molecule touches the substrate, it freezes at that site and acts as a nucleation center for subsequent aggregate growth. An aggregate initiated by a nucleation center on a column i has a domain index i. When a molecule being deposited touches a domain, it is incorporated into that domain. If a molecule being deposited touches multiple domains, we prioritize the underlying domain during assignment. Namely, the molecule belongs to the underlying domain if there exists one. However, if no underlying domain exists, the molecule is incorporated into the left or right domain depending on the ratio of the number of neighboring domain molecules. As a result of this domain assignment, we get domains in the form of trees [11]. While domains are growing, they extend laterally to capture more molecules being deposited vertically. Thus, even though several domains are initially formed, they compete with each other for capturing molecules being deposited and accordingly coarsen as shown in Fig. 1(a). The population number N of domains decreases as height h increases. The population density of the domain, $\rho = N/L$, is found to decrease by a power law as $\rho \sim h^{-0.66\pm0.03}$, as shown in Fig. 1(b).

The scaling behavior of the population density ρ can be elucidated as follows. A domain is bounded by a domain boundary on each side. When these two boundaries come in contact, the supply of molecules to the domain is cut and the domain stops growing. Therefore, domain coarsening is governed by fluctuation of the domain boundary. The fluctuation of the boundary, in turn, is related to the lateral correlation at the growth front. The scaling relation in Eq. (1) of the growth front width W indicates that the lateral correlation extends to a distance $h^{1/z}$ at a height h. Since the self-similarity of the growth front assumes no other characteristic length than the lateral correlation length, one expects that the domain boundary fluctuates laterally by the same order $h^{1/z}$. The size dependence of domain height h and domain width w was studied previously by simulations, and the relation $w \sim h^{1/z}$ was confirmed [11]. We now apply this result to the process of domain coarsening.

If the lateral fluctuation of a domain boundary is of the same order of a domain boundary separation distance, the two boundaries come in contact and the domain between them is annihilated. For a system with a domain density ρ, the average separation distance between domain boundaries is $1/\rho$. Then, domain annihilation occurs when the height of the BD aggregate increases by an annihilation height h_a, where $h_a^{1/z} \sim 1/\rho$ or $h_a \sim \rho^{-z}$. Assume that $N(h)$ domains exist at a domain height h. Then, in the height interval Δh, each domain boundary touches the other boundaries $\Delta h/h_a$ times, and the number of domains decreases as

$$N(h + \Delta h) = N(h) - N(h)\Delta h/h_a. \tag{2}$$

In terms of the domain population density $\rho(h) = N(h)/L$, the rate equation in the limit $\Delta h \to 0$ is written as

$$\frac{d\rho}{dh} = -A\rho^{1+z}. \tag{3}$$

A simple solution

$$\rho \sim h^{-1/z} \tag{4}$$

indicates that the domain population decreases with a power $1/z$. As for the dynamical scaling behavior, the BD model in one dimension is known to belong to the Kardar-Parisi-Zhang (KPZ) universality class [8,12] with the following scaling exponents: $\alpha = 1/2$, $\beta = 1/3$, and $z = 3/2$. The exponent $1/z = 2/3$ is in quite good agreement with the value 0.66 ± 0.03 obtained by fitting the data of the KMC simulation shown in Fig. 1(b). This exponent value is obtained in the asymptotic region $400 \leqslant h \leqslant 6400$. (In an early stage of $1 \leqslant h \leqslant 20$, the exponent is equal to 0.5.) The same exponent $1/z = 2/3$ was obtained previously [14] for the Eden model [13], which is also known to belong to the KPZ universality class.

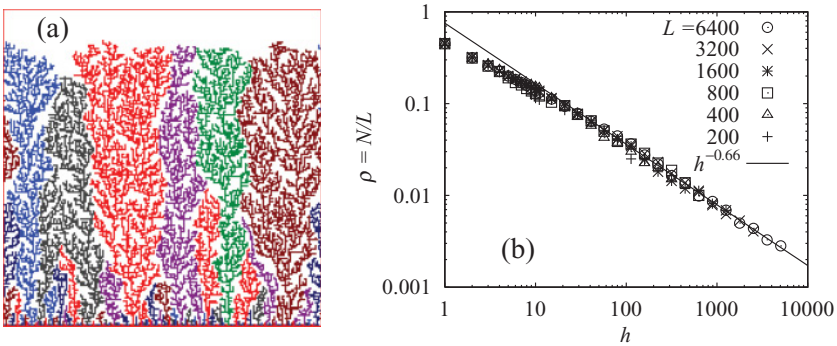

FIG. 1. (Color online) (a) Domain coarsening during BD without surface diffusion on a flat substrate. The system size is as small as $L \times H = 200^2$ for the purpose of visualization. (b) Domain population density ρ versus height h.

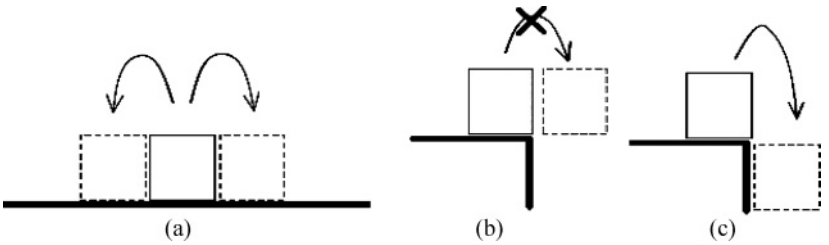

FIG. 2. (a) Intralayer diffusion with a diffusion constant D, (b) forbidden jump, and (c) intralayer transport with an additional Ehrlich-Schwoebel factor P_{ES}.

B. Ballistic deposition with surface diffusion

In several cases, the material deposited on a substrate forms a compact overlayer with a high packing density, and the growth front is flat and smooth. In that respect, a ballistically deposited aggregate with numerous voids and a self-affine growth front is far from the usual systems under investigations. To rectify this discrepancy, we incorporate a surface diffusion process on an adsorbate overlayer. It has been stated that surface diffusion affects the exponents in a random deposition model but not in the BD model [15,16]. However, another study has revealed that as diffusion proceeds, the growth mode changes from porous aggregate to compact aggregate with a smooth growth front and high packing density [10]. Then, one may expect a different type of domain coarsening.

In the present model, diffusion is mimicked such that an isolated adsorbate molecule diffuses laterally on an adsorbate layer but not on a substrate surface. Here, an isolated molecule refers to a molecule that has only one bond with an underlying adsorbate overlayer without any lateral bonds [Fig. 2(a)]. When two molecules are connected by lateral bonds, they are frozen to form an aggregate and remain stationary thereafter. An isolated molecule can jump to the left or right column at the same height level if there is at least one nearest-neighbor molecule after the diffusion jump, as shown in Fig. 2(a). If an isolated molecule is at the edge of a step, as shown in Fig. 2(b), it cannot jump laterally at the same level, but we allow it to hop down if possible, as shown in Fig. 2(c). We call the motion in Fig. 2(a) intralayer diffusion and that in Fig. 2(c) interlayer transport. Interlayer transport is introduced to make the adsorbate layer dense and smooth. Further diffusion of an adsorbate molecule along the aggregate periphery makes the aggregate more compact and dense; such diffusion is important in the asymptotic relaxation of the shape to the equilibrium state. However, in a transient process involving growth of the adsorbate island, surface smoothing is already induced by the interlayer diffusion shown in Fig. 2(c). Therefore, we neglect further shape relaxation; the adsorbate molecule is pinned to the side domains by the lateral bond after the interlayer diffusion. We may alternatively suppose that the adsorbate is anisotropic such that the lateral bond is stronger than the vertical one.

For simulations it is necessary to define various processes quantitatively. A molecule being deposited descends on a column with a deposition flux f per unit time and area. Adsorbed molecules are frozen on a substrate at their deposition sites. An isolated adsorbate molecule on an adsorbate overlayer makes a diffusion jump to the left or right at the same height level with a diffusion constant D. If an adsorbate molecule is at the edge of a step, it is usually necessary to consider the additional energy barrier that should be overcome for a molecule to move around the corner, the so-called Ehrlich-Schwoebel barrier. Thus, the rate of this interlayer transport is smaller than the intralayer diffusion D by a factor $P_{ES} \leqslant 1$.

To focus on the effect of diffusion on domain coarsening, we study an extreme case with a fast intralayer diffusion $2D/fa^4 = 10^4$ without any Ehrlich-Schwoebel barrier (i.e., $P_{ES} = 1$). After deposition and diffusion on a flat substrate, the resulting aggregate is densely packed and the growth front is flat [Figs. 3(a) and 3(b)], though some voids still exist. In Fig. 3(a), domains are differentiated by different colors (or grayscales), but since there are so many domains, two different domains that touch might have the same color (or grayscales) and are indistinguishable. Therefore, domain boundaries are depicted in Fig. 3(b) complementarily. Since an underlying domain is given priority during domain assignment, the initial domains are straight with vertical boundaries, and their population density ρ is close to unity when the domains are short, $h < 10$, as shown in Fig. 3(c). Then, domains gradually decrease in number through competition. Because the growth front is quite flat, most of the domains have the

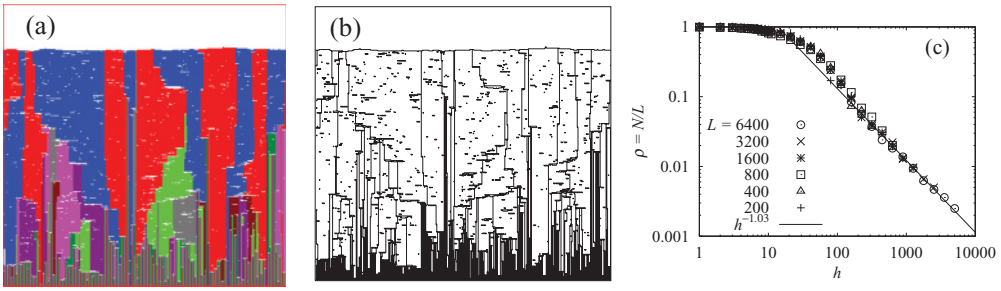

FIG. 3. (Color online) (a) Domain coarsening and (b) domain boundaries during BD with diffusion on a flat substrate. Parameters are $2D/fa^4 = 10^4$ and $P_{ES} = 1$. The system size is $L \times H = 400^2$. (c) Domain population density ρ versus height h.

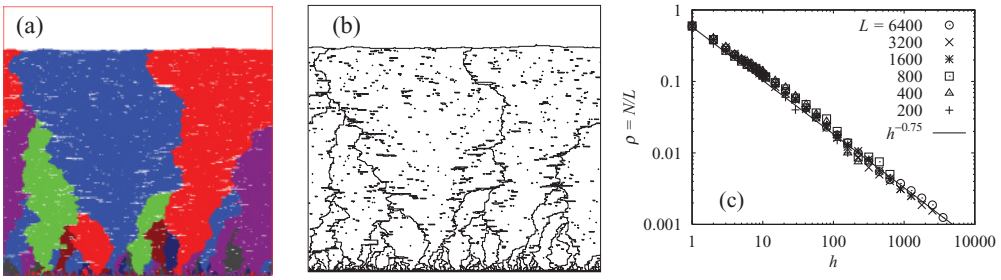

FIG. 4. (Color online) (a) Domain coarsening and (b) domain boundaries of the same BD aggregate as that in Fig. 3 but using a different procedure for assignment of domains. (c) Domain population density ρ versus height h.

same height. Therefore, when one domain grows taller than its neighboring domain, it can spread laterally over many domains. Thus, the population density decreases rapidly for $10 < h < 100$, as shown in Fig. 3(c). After this crossover, the domain density ρ for $400 \leqslant h \leqslant 6400$ seems to decrease by a power law to the height h as $\rho \sim h^{-1.01\pm0.03}$, as shown in Fig. 3(c). However, we currently do not have any analytical theory to elucidate the exponent.

In fact, the scaling behavior of the population density depends on the definition of the domain. If we alter the domain assignment such that all the neighboring domains of a molecule being deposited have equal priority rather than the underlying domain having topmost priority, domain boundaries fluctuate widely as shown in Figs. 4(a) and 4(b): The aggregate itself has the same structure as that in Figs. 3(a) and 3(b), including the configuration of the growth front, but only the domain assignment is altered. In this case, the population density of domains follows the scaling relation $\rho \sim h^{-0.75\pm0.05}$, as shown in Fig. 4(c). The system with widely fluctuating domain boundaries in Fig. 4 has a lower population density ρ than that in Fig. 3 at the same height level, but the exponent of the former is small as 0.75.

III. BALLISTIC DEPOSITION ON A PATTERNED SUBSTRATE

We now consider BD on a substrate patterned with an array of pillars, since our simulation study may be relevant to recent experimental studies on heteroepitaxial crystal growth on nanopatterned substrates [1,2,4–7], Let a pillar have a height h_p, width ℓ_p, and a periodic arrangement with periodicity λ. For simplicity, the pillar width is kept at unity in the subsequent discussion, that is, $\ell_p = 1$.

A. Ballistic deposition on a patterned substrate without diffusion

We first study domain competition during BD growth in the absence of adsorbate diffusion. Then, each domain takes the form of an irregular dendritic tree. During BD, those domains that started growing from pillar tops have an advantage over those that started growing from the bottoms of gaps between pillars; the former domains remain at a higher level than the latter, as shown in Figs. 5(a) and 5(b). If a pair of domains that started growing from neighboring pillar tops envelops those that started growing in the gap, as shown in Fig. 5(a), the latter cannot survive: There remain only those domains that started growing from pillar tops, which are almost separate from the substrate because they touch the substrate only at their roots. Therefore, the aggregate on a substrate with nanopillars is less affected by a misfit strain than that on a flat substrate.

Nanopillars aid the initial selection of domains growing from the pillar tops over those growing from gaps. However, if the pillar periodicity is large as $\lambda = 100 = 5h_p$ as shown in Fig. 5(b), some domains growing in gaps pass through the covers extending from neighboring pillar tops. Once domains growing from gaps pass through the covers, domain competition as described in Sec. II occurs. Therefore, for selection of only those domains that grow from pillar tops, the pillars should be tall and their periodicity should be small. We aim to determine the maximum periodicity λ_c of nanopillars for domain selection at a given pillar height h_p.

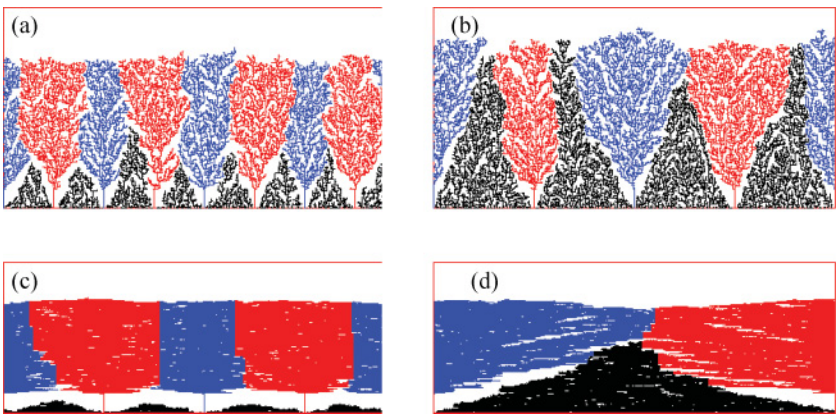

FIG. 5. (Color online) Domain competition during BD on a substrate with periodic arrays of pillars; (a) and (b) without diffusion, and (c) and (d) with diffusion. Box width is $L = 400$ and pillar height is $h_p = 20$. Pillar periodicity λ is (a) 50, (b) and (c) 100, and (d) 400, and box height is $H = 200$ for (a) and (b), and $H = 150$ for (c) and (d).

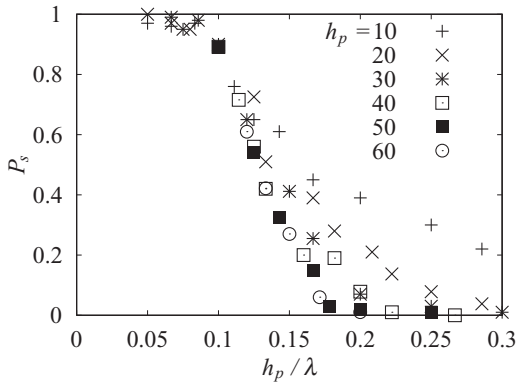

FIG. 6. Survival probability P_s of domains growing in the gap.

To quantify the selection process, we introduce a survival probability P_s. We count the number of gaps from which growing domains survive at a certain height, and we take its ratio to the total number of gaps as survival probability P_s. If all the domains growing from gaps cease to exist, P_s vanishes. On the contrary, when some of them survive, P_s takes a finite nonzero value. The probability P_s should be measured at a height neither too low nor too high: If it is measured at a very small height, domains growing in gaps are not yet covered by those growing on pillar tops; if it is measured at a very large height, the usual domain competition occurs and domains growing from the gap that have passed through the narrow channel may be suppressed by competition. Therefore, we estimated the survival probability P_s at a height of $h = 2\lambda + h_p$, where h_p is the pillar height and λ is the periodicity. The resulting P_s is plotted against the inverse periodicity h_p/λ for various pillar heights h_p in Fig. 6. As expected, domains in gaps die out when the pillars are tall enough (high h_p) or arranged close enough (small λ). On the contrary, when the pillars are far apart, domains in gaps pass through covers extending from neighboring pillar tops, and the survival probability P_s remains finite. For short pillars, the survival probability is strongly affected by a finite-size fluctuation, but as the pillar height increases, the maximum value of periodicity required for survival seems to converge to a critical value, h_p/λ_c, close to 0.2.

In order to understand the change in the survival probability P_s, we study the initial stage of domain competition shown in Fig. 5(a) more closely. We notice that a domain growing from a pillar top expands laterally as a single isolated tree until it hits domains growing from neighboring pillar tops. In fact, a single isolated domain takes the form of a tree, as shown in Fig. 7(a). The lower edge of a single tree makes a slope angle θ from the x axis; the aspect ratio of the tree is defined as $r = h/w$, where h is the height of the tree and w is its lateral width. By averaging data of many single trees in a system with a large size $L \times H = 800^2$, we obtain a slope angle $\theta = 55.6^\circ$ and an aspect ratio $r = 1.03$. With a simple assumption that an isolated domain grows in the shape of a fan with a slope angle θ and aspect ratio r, as depicted in Fig. 8, we can estimate the value of the critical periodicity λ_c as follows. When a domain growing on a pillar top extends laterally to half of the periodicity $w/2 = \lambda/2$, a pair of domains growing from neighboring pillar tops covers the gap between them. For a pillar with height h_p, two domains touch at a height $h = h_p + (\lambda/2)\tan\theta$ from the gap bottom. If this position is higher than the maximum height $h = r\lambda$ of the domains in the gap, they cannot pass through the cover formed by the lateral extension of two domains growing from pillar tops. Thus, the maximum periodicity λ_c is expected to satisfy the relation

$$h_p + \frac{\lambda_c}{2}\tan\theta = r\lambda_c \tag{5}$$

or

$$\frac{h_p}{\lambda_c} = r - \frac{1}{2}\tan\theta. \tag{6}$$

With $\theta = 55.6^\circ$ and $r = 1.03$, the calculated critical ratio $h_p/\lambda_c = 0.30$ is of the order of the simulation result 0.2, as shown in Fig. 6. The large discrepancy that remains may be due to the fluctuation, since the domain growing in the gap can survive if it accidentally passes through the narrow channel formed by the two domains from the neighboring pillar tops.

B. Ballistic deposition on a patterned substrate with diffusion

We now consider the effect of diffusion on domain selection on a patterned substrate. From the argument in the previous subsection, it can be said that initial domain selection depends on the shape of a single isolated BD aggregate. It changes as the diffusion constant D increases, as shown in Fig. 7; a single isolated domain becomes short and wide with a small slope angle θ and small aspect ratio r. For example, at $2D/fa^4 = 10^4$ in Fig. 7(d), the average slope angle is about $\theta = 7.1^\circ$, and the aspect ratio is $r = 0.11$. This data leads to the maximum periodicity of pillars $\lambda_c \approx 21h_p$, which is

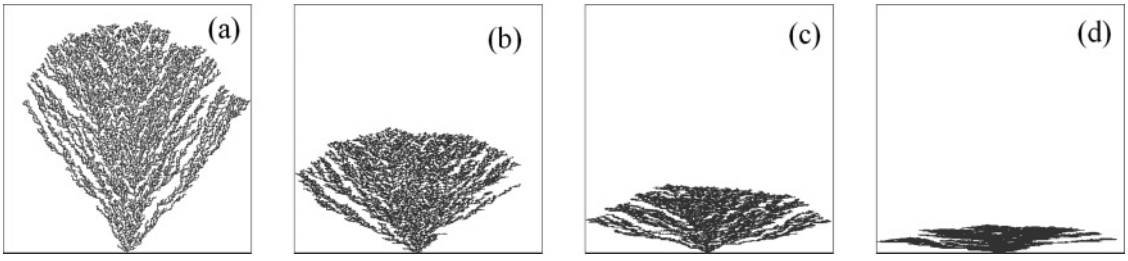

FIG. 7. Single isolated BD aggregate at various diffusion constants: (a) $2D/fa^4 = 0$, (b) 10, (c) 10^2, and (d) 10^4. The simulation box is of size $L \times H = 400^2$. The average slope angle θ and the aspect ratio r are calculated as (a) 56° and 1.03, (b) 39° and 0.61, (c) 21° and 0.32, and (d) 7.1° and 0.11, respectively.

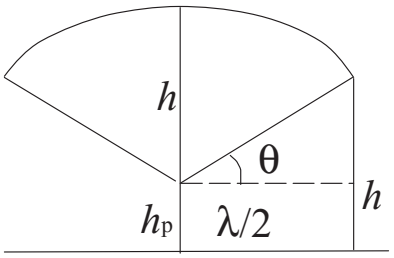

FIG. 8. Schematics of a single BD domain on a pillar.

larger than the value obtained for the case without diffusion. Actually, at a large pillar periodicity $\lambda = 100 = 5h_p$, domains growing on pillars suppress the growth of those from gaps very strongly when the diffusion is included, as shown in Fig. 5(c); this is in contrast to the survival of gap domains when diffusion is absent [Fig. 5(b)]. After domains growing from pillar tops completely suppress those growing from gaps, the domain competition discussed in Sec. II sets in among those surviving domains.

The case of a very large periodicity is shown in Fig. 5(d). Here, the periodicity is $\lambda = 400 = 20h_p$, close to the estimated limit $\lambda_c \approx 21h_p$ of the initial domain selection. A domain growing from the gap is still suppressed by neighboring ones growing from pillar tops. Surface diffusion is thus found to enhance the critical periodicity λ_c to subsequently suppress the domain growth from gaps.

IV. SUMMARY AND DISCUSSION

Coarsening of domains during ballistic deposition on a one-dimensional substrate was studied by kinetic Monte Carlo simulations. Since adsorbate molecules are supplied from above, a domain grows taller by extending its branches laterally and suppresses the growth of neighboring shorter domains. For a ballistic deposition on a flat substrate without adsorbate diffusion, the population density of domains, ρ, is found to decrease with an increase in the aggregate's height h by a power law as $\rho \sim h^{-2/3}$. When surface diffusion is introduced on an adsorbate overlayer, the grown aggregate is densely packed and domain boundaries are straight. The population density remains constant when the average height is low, but it decreases rapidly as $\rho \sim 1/h$ when the height increases.

On a substrate patterned with an array of nanopillars, domains growing from pillar tops have an advantage over those growing in gaps between pillars. When pillars are closely spaced, domains from the tops of neighboring pairs of pillars envelop those in the gap, and only those domains growing from pillar tops that are separate from the substrate remain. As the pillar periodicity λ increases or the pillar height h_p reduces, domains from the gap are able to pass through the covers and survive. A certain critical periodicity λ_c is roughly estimated from the structure of a single isolated BD domain, characterized by a slope angle θ of its lower edges and the aspect ratio r of its height to width, as in Eq. (6). Simulation results of the survival probability P_s indicate that the theoretical estimation gives the correct order of magnitude, but a more detailed analysis is necessary to account for the fluctuation effect.

Diffusion on a patterned substrate enhances the critical periodicity λ_c to suppress gap domains. After domains growing from neighboring pillar tops come in contact, they form a rather straight boundary, similar to what is observed in the heteroepitaxial experiment [5]. Since the surviving domains are located on top of the pillars, they are not very affected by the substrate and remain free from misfit strains. Thus, an overlayer on nanopillars is expected to have less defects than that grown directly on a flat substrate.

Several unresolved features still remain, especially with regard to domain coarsening with surface diffusion: The scaling behavior of the population density with an exponent ~ 1 is waiting for explanation. Surface diffusion is known to have various effects on the growth front, such as anomalous scaling [17,18] or crossover phenomena [19–21]. The change in the growth front certainly affects the domain structure, but it is not the only factor controlling the domain growth presented here. The process of domain assignment also influences the scaling exponents, as explained in Sec. II. Therefore, further studies are necessary to investigate this.

Further, a generic method for coarsening has recently been proposed [22] in the case that the evolution equation of the front is known and its nonlinear steady state is available. For the present problem of domain coarsening, we need the evolution equation of the population density. In the absence of diffusion, this is Eq. (3). In the presence of diffusion, a proper evolution equation remains to be determined.

The model considered here is a very simplified one, and it needs to be modified to be more realistic. Instead of diffusion of an isolated adsorbate molecule, diffusion should be a thermally activated process at the cost of breaking chemical bonds. For adsorbate crystal with several facet faces, not only is a nearest-neighbor interaction relevant but also further neighbor interactions have to be taken into account [23]. Extension to a three-dimensional system with a two-dimensional substrate is also necessary. Another interesting case to consider is when nanopillars have some variation in height. Then, the domains on the highest pillars dominate the others on lower pillars. When the number of surviving domains is reduced, the adsorbate overlayer becomes homogenous. This situation might correspond to experiments on buffer layers [1,2], and it seems worthy of investigation in the future.

ACKNOWLEDGMENT

Y.S. acknowledges the financial support provided by a Grant-in-Aid for Scientific Research, Grant Nos. 19540410 and 23540456, from the Japan Society for the Promotion of Science.

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