APPLIED PHYSICS LETTERS **94**, 153110 (2009)

Scaling of the surface migration length in nanoscale patterned growth

S. C. Lee[a)] and S. R. J. Brueck
Center for High Technology Materials, Department of Electrical and Computer Engineering, University of New Mexico, 1313 Goddard SE, Albuquerque, New Mexico 87106, USA

(Received 23 January 2009; accepted 23 March 2009; published online 16 April 2009)

Scaling of the surface migration length in nanoscale patterned growth (NPG) is investigated as a function of the lateral dimension L_M of a mask film fabricated on a substrate for selective epitaxy. By reducing L_M below the surface migration length, any nucleation on the mask is avoided through the evaporation and surface out-diffusion of adatoms. The upper limit of L_M for NPG $L_{M,c}$ corresponds to the surface migration length on the mask. An equation, identical to that for two-dimensional step-flow growth, is derived for NPG. However, the boundary conditions at the substrate-mask interface are affected by the surface potential difference and are different from those at the terrace edges of a homogeneous stepped surface. This results in a scaling law for surface migration length, which is proportional to the diffusion constant D and the critical incident flux F_c in the form $(D/F_c)^{1/\alpha}$ with α decreasing from 4 to 2 as evaporation becomes dominant. NPG of GaAs for $L_{M,c}\sim 200$ nm($\alpha\sim 3.8$) is demonstrated at ~600 °C with molecular beam epitaxy. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3117366]

Nanoscale patterned growth (NPG) provides various growth modes which are unavailable in macroscale epitaxy.[1] One of them is selective epitaxy. The basic idea is to reduce the lateral dimension L_M of a masked substrate area below the surface migration length of an adatom, defined as its average migration distance before evaporation or interaction with other atoms for clustering, so that they can be removed from the masked area without nucleation. Then, any nucleation is avoided on the masked area resulting in selective epitaxy. NPG is significant for the epitaxial growth of customized nanostructures such as patterned quantum dots[2] and the study of the nucleation in thin film growth, the major focus of this article.

Extensive theoretical and experimental studies of thin film growth relying on step-flow growth have been reported.[3] Most of these articles, however, relate to extremely low deposition rates or submonolayer depositions at low growth temperatures and are not applicable to practical epitaxial growth.[3–6] In this report, we examine the nucleation of thin film growth with NPG under typical epitaxy conditions, very different from those of the reported work. The model for NPG is identical to that for step-flow growth of critical cluster size $i=1$ in two dimension (2D) if L_M is regarded as the terrace length of a step. However, the boundary conditions of such a heterogeneous patterned surface are different from those of a homogeneous stepped surface, and as a result, a different scaling for the surface migration length is derived. This scaling is deduced from the NPG equation and compared with those from other models. To confirm the validity of the scaling, NPG is experimentally demonstrated using molecular beam epitaxy (MBE).

As seen in Fig. 1(a), NPG is modeled by deposition of GaAs onto a heterogeneous substrate consisting of a circular SiO_2 disk of diameter $L_M(=2r)$ fabricated atop a GaAs substrate. The basic requirement for NPG (evaporation of an adatom from the SiO_2 disk or surface out-diffusion from it to an adjacent substrate surface) is similar to that for step-flow growth where an atom incident on a terrace should migrate to a step edge for incorporation into an epilayer. Thus, the equation for NPG is the same as that for step-flow growth, given by[3]

$$\frac{\partial n}{\partial t}=F-\frac{n}{\tau_{\mathrm{evap}}}+D\nabla^2 n, \qquad (1)$$

where n is the number of Ga adatoms per unit area of the SiO_2 disk at time t; F is the Ga flux incident on a unit SiO_2 area per unit time; $\tau_{\mathrm{evap}}=\nu_0^{-1}\exp(E_{\mathrm{des}}/k_BT)$ is the evaporation-limited surface residence time of Ga on the SiO_2 which is described with a desorption rate constant ν_0, activation energy of Ga desorption from SiO_2, E_{des}, Boltzmann constant k_B, and growth temperature T; $D=\nu_d\exp(-E_{\mathrm{diff}}/k_BT)$ is the diffusion constant characterized by an activation energy for surface diffusion of Ga on SiO_2, E_{diff}, and a proportionality constant ν_d. Equation (1) keeping

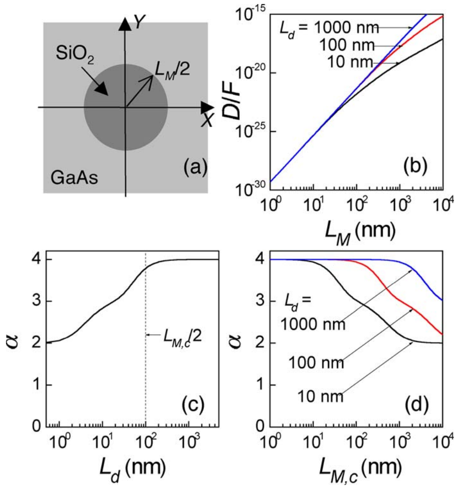

FIG. 1. (Color online) (a) A schematic illustration of a SiO_2 disk of diameter L_M fabricated on a GaAs substrate. (b) A plot of L_M vs D/F of Eq. (2) for three different L_d's. (c) A plot of L_d vs α for $L_{M,c}=200$ nm. The dashed line indicates $2L_d=L_{M,c}=200$ nm from (a) for $\alpha=3.8$. (d) A plot of $L_{M,c}$ vs α obtained from (b) with the approximation of Eq. (2).

[a)] Electronic mail: sclee@chtm.unm.edu.

the evaporation term (n/τ_{evap}) is applicable at any growth temperature. The surface potential energy changes more significantly at the heterogeneous substrate-mask boundary $[r=L_M/2$ in Fig. 1(a)] than the homogeneous terrace edge on a stepped surface. As shown in previous studies of NPG,[1,7] the surface potential becomes lower on the GaAs substrate and the negative gradient near the boundary plays the role of a driving force for out-diffusion of adatoms from the SiO_2 disk to the substrate. Then, the appropriate boundary condition is $n(L_M/2,t)\sim 0$ and the reverse diffusion flux from GaAs onto SiO_2 is relatively insignificant. These boundary conditions are very important since they result in substantial differences from previous studies. Applying them to Eq. (1) and integrating n over the SiO_2 disk, the steady-state solution to Eq. (1) can be written as[8]

$$\frac{D}{F} = \frac{\pi L_M{}^4}{4N}\sum_{m=1}^{\infty}[z_m{}^2 + (L_M/2L_d)^2]^{-1}; \quad L_{M,c} \sim \left(\frac{D}{F_c}\right)^{1/\alpha}. \tag{2}$$

Here, $N=\int_A n da$ is the total number of Ga atoms on the SiO_2 disk, z_m is the mth zero of the zeroth order Bessel function, $L_d=\sqrt{\tau_{\text{evap}}D}$ is the diffusion length of a Ga atom on unpatterned SiO_2, and α is a dimensionless exponent. In Eq. (2), $L_{M,c}$ is the upper limit of L_M for NPG and can be interpreted as the surface migration length of a Ga adatom on SiO_2 at a given T, and F_c is the critical flux for maintaining NPG. To suppress nucleation on the SiO_2 disk, F must be less than F_c for a given $L_{M,c}$. Then, Eq. (2) provides the relation between F_c and $L_{M,c}$ for NPG and is referred to as the NPG equation. For selective growth, N can only be at most a few regardless of $L_{M,c}$. Then, we can set $N=1$ in Eq. (2). This does not affect the calculation of α. Figure 1(b) shows a plot of D/F versus L_M for $L_d=10$ nm, 100 nm, and 1 μm at a given T. Also, Fig. 1(d) presents a plot of $L_{M,c}$ versus α obtained from Fig. 1(b) within the approximation of Eq. (2).

As seen in Fig. 1(d), α decreases from 4 as $L_{M,c}$ increases. Two limiting cases are (a) $L_M \gg L_d(\alpha\rightarrow 2)$ and (b) $L_M \ll L_d(\alpha=4)$. Case (a) has been discussed previously;[7] nucleation-free growth on SiO_2 is achieved only by evaporation and this condition leads to $F_c \propto \exp[-(2E_{\text{des}}-E_{\text{diff}})/k_BT]$ with $E_{\text{des}}>E_{\text{diff}}/2$.[9] In case (b), the absence of nucleation on the SiO_2 disk is accomplished entirely by surface out-diffusion. In Figs. 1(b) and 1(d), all α's converge to 4 as $L_{M,c}\rightarrow 0$. Because of the different boundary conditions, the exponent for this case is different from the reported work. In Monte Carlo and other simulations, $L_d \propto (D/F)^{1/6}$ for $i=1$.[3–6] The reduction in α with evaporation is qualitatively similar to the result of other reports.[4] The surface migration length used in this work is somewhat different from but comparable to L_d.

In practical growth, both evaporation and surface out-diffusion are important for suppressing nucleation on the SiO_2. Roughly, this corresponds to an incorporation rate less than unity or partial evaporation. As seen in Fig. 1(d), $L_{M,c} \propto (D/F_c)^{1/\alpha}$ with α that decreases below 4 with $L_{M,c}$. This variation is due to evaporation and has not been intensively studied. This is the important result we focus on in this work and will be discussed along with the experimental data.

To examine NPG experimentally, 2D arrays of circular holes and posts were fabricated into a ~30-nm-thick SiO_2 film atop a GaAs(001) substrate by interferometric lithography and dry etching.[1] The pattern period ranged from 260 to 350 nm. GaAs was deposited by MBE onto these nanopatterned samples. Figure 2 shows scanning electron microscope (SEM) images taken from an as-grown sample at $T=570$ °C with a Ga flux of $F=0.33\times 10^{14}$ atoms/cm^2 s that show how $L_{M,c}$ was measured experimentally. In Fig. 2, L_M was reduced from (a) infinity to (f) ~70 nm. We approximate these SiO_2 masks of Figs. 2(e) and 2(f) as circular disks for comparison with Eq. (2). The six different L_M's presented in Fig. 2 were realized on a single substrate by controlling dry etch time with a shadow mask to minimize the pattern-to-pattern variation in T and F in the $L_{M,c}$ measurement. The growth conditions and deposition amounts are given in Table I.

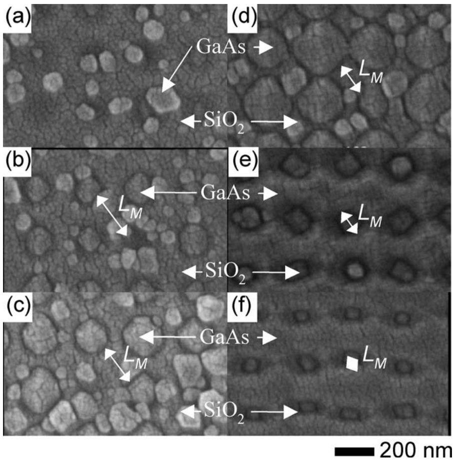

FIG. 2. Top-view SEM images of the as-grown sample at $T=570$ °C having (a) L_M of infinity (no pattern), (b) ~270 nm, (c) ~220 nm, (d) ~180 nm, (e) ~120 nm, and (f) ~70 nm. In (f), the alignment and shape of SiO_2 masks are slightly degraded because of the fluctuation of dry etching at such small scales. It should be noted that the periodic circular shapes in (e) and (f) are SiO_2, while those in (b) through (d) are GaAs.

Figure 2 shows the transition to selective epitaxy in NPG. Nucleation on the SiO_2 disks begins to reduce for (d) $L_M\sim 180$ nm and disappears entirely for (e) $L_M \leq 120$ nm. Thus, $L_{M,c}\sim 120$ nm for the given T and F. This measurement of surface migration length is very different from previous experiments typically performed by scanning tunneling microscopy with submonolayer deposition. The measured $L_{M,c}$'s are considerably smaller than the micrometer-scale surface migration length of Ga on GaAs(001) for this temperature range.[10] From Table I, $L_{M,c}$ is reduced as both T and F are decreased.

In Fig. 2(a), the partial coverage of GaAs on the wide, unpatterned SiO_2 surface shows that the Ga incorporation rate is less than unity, and therefore $\alpha<4$. Other samples also show similar coverage for the deposition rates summarized in Table I. All of the $L_{M,c}$'s in Table I are around 200 nm and the same α can be assumed from Fig. 1(d). Figure 1(c) shows a plot of L_d versus α for $L_{M,c}=200$ nm from Eq.

TABLE I. Summary of NPG conditions.

Growth temperature (°C)	570	600	630
Ga flux (10^{14}/cm^2 s)	0.33	0.65	1.6
Deposition amount (nm/cm^2)	50	100	30
Pattern period (nm)	260	350	330
$L_{M,c}$ (nm)	90–150	170–220	230–290

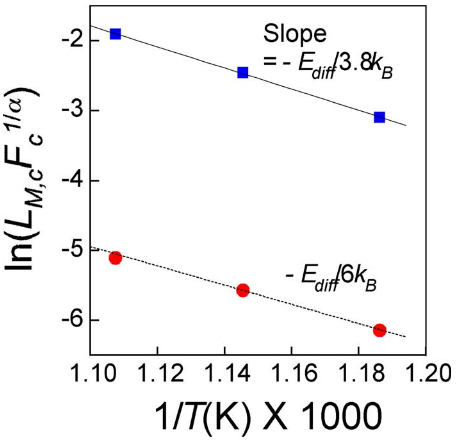

FIG. 3. (Color online) A plot of $L_{M,c}F_c^{\,1/\alpha}$ vs $1/T$ from Table I with $\alpha = 3.8$ (square) and 6 (circle). For each measured $L_{M,c}$ at given T, the corresponding Ga flux in Table I becomes F_c. In the plot, the median for each $L_{M,c}$ in Table I was used.

(2). From this figure, $\alpha \sim 3.8$ is a good approximation for the NPG in Table I. This $\alpha < 4$ assigned from the experimental $L_{M,c}$'s not only includes the contribution of evaporation to NPG consistent with the partial coverage in Fig. 2(a) but also explains the nucleation-free areas of the SiO_2 disks assisted by surface out-diffusion in Figs. 2(e) and 2(f).

Figure 3 shows an Arrhenius plot of $L_{M,c}F_c^{\,1/\alpha}(\sim D^{1/\alpha})$ versus $1/T$ obtained from Table I with $\alpha = 3.8$ (squares) and $\alpha = 6$ (circles). From Fig. 3, E_{diff} is approximately 4.9 eV when $\alpha = 3.8$. This E_{diff} is significantly greater than the activation energy of surface diffusion for a Ga adatom on a GaAs surface ~ 1 eV.[11] This suggests that a Ga adatom is comparatively immobile on a SiO_2 surface and is consistent with the short $L_{M,c}$'s observed experimentally. It should be noted that the E_{diff} of this work is valid around 600 °C. The high E_{diff} obtained with $\alpha \sim 3.8$ does not conflict with previous reports. E_{des} for a Ga atom on an SiO_2 surface in Eq. (2) must be lower than 4.7 eV, the activation energy for Ga desorption from a GaAs surface,[12] since selective epitaxy of GaAs on a SiO_2-patterned substrate relying solely on evaporation has been demonstrated.[7] Then, from the relation of $E_{\text{des}} > E_{\text{diff}}/2$, E_{des} should be greater than ~ 2.5 eV, which is consistent with the upper limit of 4.7 eV. E_{diff} calculated with $\alpha = 6$ gives ~ 6.8 eV which does not allow a realistic margin for E_{des} in consideration of the approximation applied in this work. For this reason, $\alpha = 3.8$ is more plausible than $\alpha = 6$ in analyzing our data and indirectly supports Eq. (2). Thus, the model and equation for NPG suggest an ultimate way to measure surface migration length and our data provide experimental evidence quantifying its scaling exponent with the inclusion of evaporation.

In conclusion, NPG assisted by evaporation and surface out-diffusion has been proposed and demonstrated. The surface migration length of an adatom on a mask film $L_{M,c}$ for NPG is proportional to $(D/F)^{1/\alpha}$ with $2 < \alpha \leq 4$. As evaporation becomes dominant, α decreases from 4 with increasing $L_{M,c}$. In the experiment of GaAs growth on a SiO_2-patterned substrate with partial evaporation, $\alpha = 3.8$ for $L_{M,c} \sim 200$ nm of a Ga adatom on a SiO_2 disk has been measured at ~ 600 °C along with the equation.

This work was supported by DoE/Basic Energy Sciences under subcontract #50615 from Sandia National Laboratories.

[1] S. C. Lee, K. J. Malloy, and S. R. J. Brueck, J. Appl. Phys. **90**, 4163 (2001).
[2] S. Birudavolu, N. Nuntawong, G. Balakrishnan, Y. C. Xin, S. Huang, S. C. Lee, S. R. J. Brueck, C. P. Hains, and D. L. Huffaker, Appl. Phys. Lett. **85**, 2337 (2004).
[3] A. Pimpinelli and J. Villian, *Physics of Crystal Growth* (Cambridge University Press, Cambridge, 1998), and references therein.
[4] J. A. Venables, G. D. T. Spiller, and M. Hanbuecken, Rep. Prog. Phys. **47**, 399 (1984).
[5] M. C. Bartelt and J. W. Evans, Phys. Rev. B **46**, 12675 (1992).
[6] M. Schroeder and D. E. Wolf, Phys. Rev. Lett. **74**, 2062 (1995).
[7] S. C. Lee, L. R. Dawson, S. R. J. Brueck, and A. Stintz, J. Appl. Phys. **96**, 4856 (2004).
[8] R. V. Churchill and J. W. Brown, *Fourier Series and Boundary Value Problems*, 3rd ed. (McGraw-Hill, New York, 1978), p. 174.
[9] In this case, $L_{M,c}$ is physically meaningless since all Ga atoms would evaporate from the SiO_2 disk before reaching the edge of the mask. Experimentally, the corresponding α would be valid for L_M, which is at most a few times of L_d and does not approach 2 closely as $L_M \rightarrow \infty$.
[10] M. Hata, T. Isu, A. Watanabe, and Y. Katayama, J. Vac. Sci. Technol. B **8**, 692 (1990).
[11] J. H. Neave, P. J. Dobson, B. A. Joyce, and J. Zhang, Appl. Phys. Lett. **47**, 100 (1985).
[12] J. P. Reithmaier, R. F. Broom, and H. P. Meier, Appl. Phys. Lett. **61**, 1222 (1992).