ELSEVIER

Surface Science 382 (1997) 170–177

surface science

Transitions in critical size in metal (100) and metal (111) homoepitaxy

Jacques G. Amar, Fereydoon Family *

Department of Physics, Emory University, Atlanta, GA 30322, USA

Received 19 September 1996; accepted for publication 22 January 1997

Abstract

Transitions in the critical island size i in submonolayer growth from $i=1$ to $i=2$ and from $i=1$ to $i=3$, corresponding to homoepitaxial growth on metal (111) and (100) surfaces, are studied using kinetic Monte Carlo simulations of a restricted pair-bond model, both with and without island relaxation, and are compared with rate-equation predictions. In both cases, the rate equations significantly underestimate the transition temperature from $i=1$ behavior to a higher critical island size. The difference is due to the neglect of spatial correlations in the standard rate-equation approach. A partial solution involving the use of effective detachment rates is proposed. © 1997 Elsevier Science B.V.

Keywords: Epitaxy; Models of non-equilibrium phenomena; Nucleation; Surface diffusion

1. Introduction

Recently there has been a good deal of experimental [1–11] and theoretical [12–30] interest in the scaling properties of the island density and island-size distribution in submonolayer epitaxial growth. One important motivation is that the dependence of the submonolayer island density on deposition rate and temperature at fixed coverage θ in the precoalescence regime may be used to identify important activation energies for microscopic processes on the surface. One fundamental concept which has been extensively used is that of a critical island size corresponding to one less than the number of atoms in the smallest stable island [13]. For a given critical island size i, standard rate-equation theory [13] predicts that at fixed coverage, the island density N and single-adatom (monomer) density N_1 scale as

$$N \sim (D/F)^{-\chi_i} \exp\left[\frac{E_i}{(i+2)k_B T}\right], \tag{1a}$$

$$N_1 \sim (D/F)^{-(1-\chi_i)} \exp\left[\frac{-E_i}{(i+2)k_B T}\right], \tag{1b}$$

where $\chi_i = i/(i+2)$ and $R = D/F$ is the ratio of the hopping rate D ($D = D_0 \, e^{-E_a/k_B T}$) of an isolated adatom to the deposition flux F, while E_i is the binding energy of the critical cluster. From the flux dependence of the island density at fixed temperature, the exponent χ_i and the critical island size i may be determined experimentally, while the temperature dependence of the island density yields an estimate for the combination $E_i + iE_a$, where E_a is the activation energy for monomer diffusion.

* Corresponding author. Fax: +1 404 7270873; e-mail: phyff@emory.edu

PII S0039-6028(97)00121-0

In the simplest case (corresponding to $i=1$ at low temperature), one has $E_i=0$ and the activation energy for monomer diffusion E_a may be determined directly from a measurement of the temperature dependence of the island density [7].

Eqs. (1a) and (1b) may be derived from a contraction of the standard rate equations [13] using the assumption that the density N_i of critical islands satisfies the quasi-equilibrium Walton relation [12] $N_i \sim e^{E_i/k_B T} N_1^i$. However, as has been noted by a number of authors [20,23] the rate-equation approach ignores correlations. In particular, as pointed out in Ref. [26], the standard rate-equation approach assumes that a monomer which has detached from an island is equally likely to attach to any existing island rather than to a nearby island. The classical rate-equation approach also assumes that in the case of a critical island size i, all islands of a size $i+1$ or larger are stable, although for $i \neq 1$ this is unlikely for many systems, even in the case of rapid island relaxation. Nevertheless, recent arguments and kinetic Monte Carlo simulations [24,25] have shown that for more realistic models in which the smallest stable island size is given by $i+1$, but for which detachment from islands larger than $i+1$ is allowed, the flux dependence of the island density predicted by Eqs. (1a) and (1b) still holds (i.e. $\chi_i = i/(i+2)$). However, only preliminary simulation results for the dependence of the island density on the binding energy of the critical cluster have so far been reported [25–27].

In this paper we present the results of extensive kinetic Monte Carlo (KMC) simulations carried out in order to study the crossover scaling behavior of the island density as a function of temperature and critical cluster energy. Of particular interest is a comparison between simulation results for the transition from $i=1$ to $i=3$ on a square lattice (corresponding to growth on a metal (100) surface) and the rate-equation prediction (Eqs. (1a) and (1b), as well as with recent results for the crossover scaling behavior obtained by direct numerical integration of the standard rate equations [28]. We also present a comparison between simulation results for the case of a transition from $i=1$ to $i=2$ on a triangular lattice (corresponding to growth on a metal (111) surface) and the corresponding rate-equation results. We note that a transition from a critical island size of 1 at low temperature to a critical island size of 2 at higher temperature is expected to occur in metal (111) homoepitaxy [25] and is recently believed to have been observed in Rh/Rh(111) deposition [11]. In addition, a transition from $i=1$ to $i=3$ has also been reported for Fe/Fe(100) deposition at room temperature [7], and may be expected to occur in a variety of other metal (100) homoepitaxy systems. Therefore, the results of our simulations may be useful in the analysis and interpretation of a variety of experiments.

2. Simulations

In order to consider the simplest possible case, simulations were carried out using a restricted pair-bond model [25], in which the detachment rate for adatoms with one nearest-neighbor bond is given by $D_1 = r_1 D$ (where $r_1 = e^{E_N/k_B T}$ and E_N is the nearest-neighbor bond energy), while atoms with two or more nearest-neighbor bonds are assumed to be immobile. This implies [25] that for a sufficiently large rate of one-bond detachment (i.e. sufficiently large temperature), the critical island size is equal to 2 on a triangular lattice (corresponding to a stable trimer), while on a square lattice the critical island size is equal to 3 (stable tetramer). As already noted in Refs. [25–28], such a restricted pair-bond model may be appropriate for systems for which the nearest-neighbor bond energy E_N is sufficiently large that the rate of two-bond detachment is negligible. As in Ref. [25], simulations were carried out on both square lattices (corresponding to metal (100) surfaces) and triangular lattices (corresponding to metal (111) surfaces). In order to study the effects of island relaxation on the scaling behavior, simulations were carried out both with and without edge diffusion. In the case in which edge diffusion was included, a separate activation energy for diffusion of adatoms with one nearest-neighbor bond along edges and around kinks was included at a rate given by $D_e = D$.

We note that for the model considered here, the standard rate-equation prediction (Eqs. (1a) and

(1b)) implies (since $E_i=(i-1)E_N$ for $i=1, 2$ (3) on a triangular (square) lattice [25,27,29])

$$N \sim r_1^{-\phi_i} R^{-1/(i+2)} \sim R^{-1/3} (r_1^{3/2} R)^{-2(i-1)/[3(i+2)]}, \tag{2a}$$

$$N_1 \sim r_1^{\phi_i'} R^{-2/(i+2)} \sim R^{-2/3} (r_1^{3/2} R)^{2(i-1)/[3(i+2)]}, \tag{2b}$$

where $\phi_i'=\phi_i=(i-1)/(i+2)$. If we assume, as first suggested in Ref. [25], that the crossover scaling function for the island density N (at fixed coverage) from $i=1$ to $i=k$ has the general form

$$N \sim R^{-1/3} f_{1k}(r_1^{x_{1k}} R), \tag{3}$$

where x_{1k} is a crossover exponent, and where $f(u) \sim$ const. for $u \ll 1$ and $f(u) \sim u^{-2(k-1)/[3(k+2)]}$ for $u \gg 1$, then the standard rate-equation prediction (Eqs. (2a) and (2b) implies that the value of the crossover exponent is given by $x_{12}=x_{13}=3/2$ [27–29].

3. Results and discussion

Fig. 1 shows the results obtained from simulations of our restricted pair-bond model on a triangular lattice. In particular, Fig. 1a shows the crossover scaling function $f_{12}(r_1^{3/2}R)=NR^{1/3}$ (where N is the island density at fixed coverage $\theta=0.1$) for the case without edge diffusion ($D_e=0$) using the rate-equation prediction $x_{12}=3/2$. As can be seen, there is good agreement with the scaling form (Eq. (3) using the exponents $\phi_2=1/4$, $x_{12}=3/2$ predicted by Eqs. (1a) and (1b) and Eqs. (2a) and (2b). In particular, for small values of $u=r_1^{3/2}R$ one has $f_{12}(u) \sim$ const., corresponding to $i=1$ behavior, while for very large values of u one has $f_{12}(u) \sim u^{-1/6}$, corresponding to $i=2$. Thus, the standard rate-equation theory appears to give the correct scaling form in this case. We note, however, the existence of small deviations from the expected $f_{12} \sim u^{-1/6}$ behavior at large u for small D/F. This indicates that relatively large values of D/F are needed for the asymptotic scaling behavior predicted by Eqs. (1a) and (1b) and Eqs. (2a) and (2b) to be observed.

Fig. 1b shows the corresponding results for the effective scaling exponent $\chi=\mathrm{d}(\ln N)/\mathrm{d}(\ln F)$ as a function of u for both the case without edge diffusion (filled symbols, $D_e=0$) and the case of rapid island relaxation via fast edge diffusion (open symbols, $D_e=D$). In the latter case, dimer edge diffusion has been suppressed in order to prevent dimer mobility. As can be seen, the results with fast edge diffusion are significantly different from those without edge diffusion. In particular, the crossover away from $\chi=1/3$ occurs significantly earlier in the case of fast island relaxation than without island relaxation. This is most probably

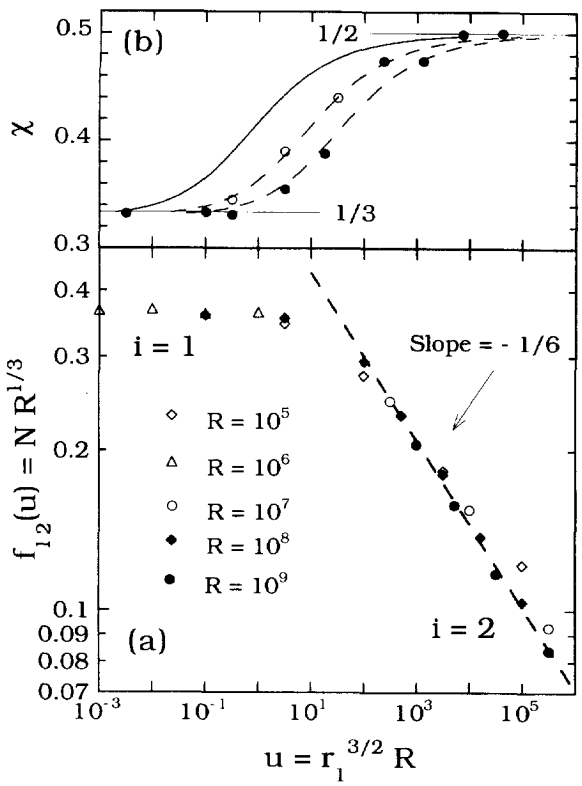

Fig. 1. Simulation and rate-equation results for the restricted pair-bond model on a triangular lattice as function of r_1 and R. (a) Island-density crossover scaling function $f_{12}(r_1 3/2R)$. (b) Scaling exponent $\chi=\mathrm{d}(\ln N)/\mathrm{d}[\ln(F)]$ as a function of crossover scaling variable $r_1^{3/2}R$. Filled symbols correspond to results with no edge diffusion, while open symbols correspond to results with rapid edge diffusion. The solid curve is the rate-equation prediction from numerical integration of Eqs. (4a), (4b) and (4c). Dashed curves are corrected rate-equation fits (see text) with $\alpha=0.33$ (through open symbols) and $\alpha=0.13$ (through filled symbols).

due to the effects of edge diffusion on the stability of small clusters. We now consider a more quantitative comparison of our results with standard rate-equation theory.

As is well known [19,20,23], the use of rate equations with constant capture cross-sections is not sufficient to provide a quantitative measure of the island density as a function of coverage, even though such an approximation may predict correctly the dependence of the island density on deposition flux for a given critical island size. However, it has been argued [28] that the standard rate-equation approach may still be used to accurately predict transitions in critical island size and crossover scaling behavior, and in particular to predict the value of the exponent $\chi = \mathrm{d}(\ln N)/\mathrm{d}(\ln F)$. In order to test this, we compare our simulation results in Fig. 1b with the results obtained from numerical integration of the contracted set of rate equations

$$\mathrm{d}N_1/\mathrm{d}\theta = 1 - 2R\sigma_1 N_1^2 - R\sigma_2 N_1 N_2 - R\sigma_{\mathrm{av}} N_1 N + 4r_1' R N_2, \quad (4a)$$

$$\mathrm{d}N_2/\mathrm{d}\theta = R\sigma_1 N_1^2 - R\sigma_2 N_1 N_2 - 2r_1' R N_2, \quad (4b)$$

$$\mathrm{d}N/\mathrm{d}\theta = R\sigma_2 N_1 N_2, \quad (4c)$$

which take into account adatom deposition and diffusion as well as dimer formation and break-up, trimer formation, and attachment of isolated adatoms to stable clusters of size greater than 2. Here, N_1, N_2, and N refer to the density of monomers, dimers, and islands of size 3 or greater, respectively, and for simplicity the capture cross-sections σ_1, σ_2 and σ_{av} are all taken to be constant, as in Ref. [28]. In Eqs. (4a), (4b) and (4c), a corrected effective detachment rate $r_1' = 0.6 r_1$ has been used, since for an adatom with one bond on a triangular lattice, only three of the five possible directions for one-bond diffusion actually correspond to detachment. We note that this set of rate equations corresponds to the case in which all islands of size 3 or greater are stable (no detachment) and is appropriate for our simulations with rapid edge-diffusion, since in this case any single-bonded adatom which is part of an island of size 3 or greater will rapidly move to a nearest-neighbor site with two or more bonds. We also note that the rate-equation results shown in Fig. 1b all correspond to $\sigma_1 = \sigma_2 = \sigma_{\mathrm{av}} = 1$. However, the rate equations were also integrated numerically using significantly larger values (up to $\sigma_{\mathrm{av}} = 7$), and very little change in the results was observed. This is not surprising, since the exponent χ is related to the effective "critical island size" i, which depends primarily on the effective detachment rate and not on the capture cross-sections.

Fig. 1b shows a comparison between our simulation results for χ with rapid edge diffusion (open symbols) and the rate equation results from Eqs. (4a), (4b) and (4c) (solid curve). As can be seen in Fig. 1b, the rate-equation prediction with $r_1' = 0.6 r_1$ (solid curve) does not agree with our rapid edge-diffusion results. While this is due in part to the assumption of constant capture cross-sections σ_s, it is also due to the fact that the rate equations do not take into account the probability that a dimer which has just broken up is more likely to reform than is one of the adatoms to attach to a nearby island. This leads to an effective detachment probability $r_{1\mathrm{eff}}'$ which is smaller than the actual detachment rate r_1'. Accordingly, we have replaced the detachment probability in Eqs. (4a), (4b) and (4c) by an effective detachment rate $r_{1\mathrm{eff}}' = \alpha r_1'$ in an effort to fit the simulation results. As can be seen by the dashed curve in Fig. 1b, reducing the actual detachment rate by a factor of about 1/3 ($\alpha \simeq 0.33$) gives a good fit to our simulation results with edge diffusion. We note, however, that a direct analysis of the probability of an isolated dimer which has just broken up to re-form gives a somewhat higher value ($\alpha \simeq 0.45$), so that a simple argument does not entirely account for this result. Also shown in Fig. 1b is a rate-equation fit to our simulation results without edge-diffusion using $\alpha = 0.13$. We note that in this case the fit is somewhat heuristic, since the fact that there is a significant probability for detachment from large clusters in the absence of edge diffusion has not been taken into account in our rate equations.

We have also checked whether the Walton relation holds in the limit of large u corresponding to $i = 2$ behavior by measuring the Walton ratio $R_{\mathrm{W}} = N_2 \, \mathrm{e}^{E_2/k_{\mathrm{B}}T}/N_1^2 = r_1 N_2/N_1^2$ in our simulations as a function of coverage for two different sets of

values of r_1 and R. As expected, we find relatively good agreement ($R_W \simeq 3 \pm 0.5$ for $0.01 \leq \theta \leq 0.1$ for $r_1 = 0.01$, $R = 10^7$ and $r_1 = 0.003$, $R = 10^8$). We note that the fluctuations in R_W are quite large due to the very small magnitude of the monomer and critical island density, especially at higher coverages. We have also checked the scaling of the monomer density N_1 as a function of r_1 in this regime, and again find relatively good agreement with Eq. (1b). Accordingly, we conclude that although the standard rate-equation approach (including the Walton relation) gives the correct scaling behavior and scaling exponents for this system in the limit of both $i = 1$ and $i = 2$, it does not give a quantitative estimate of the crossover behavior. This is due to the use of constant capture cross-sections and the neglect of correlations in detachment/attachment in the standard rate-equation approach.

We now consider the transition from $i = 1$ to $i = 3$. Figs. 2 and 3 show our results for the island density and scaling exponent $\chi = d(\ln N)/d(\ln F)$ (where N is the island density at fixed coverage $\theta = 0.1$) obtained from simulations of our restricted

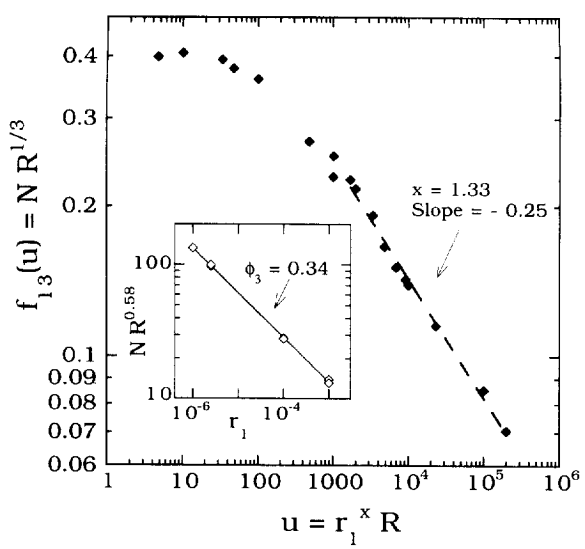

Fig. 2. Island-density crossover scaling function $f_{13}(u) = NR^{1/3}$, where $u = r_1^{x_{13}} R$ and the crossover exponent $x_{13} = 1.33$ are obtained from simulations of the restricted pair-bond model on a square lattice. The fit for large u (dashed line) has a slope of -0.25. The inset shows the scaling behavior of island density N as a function of the detachment rate r_1 in the $i = 3$ regime.

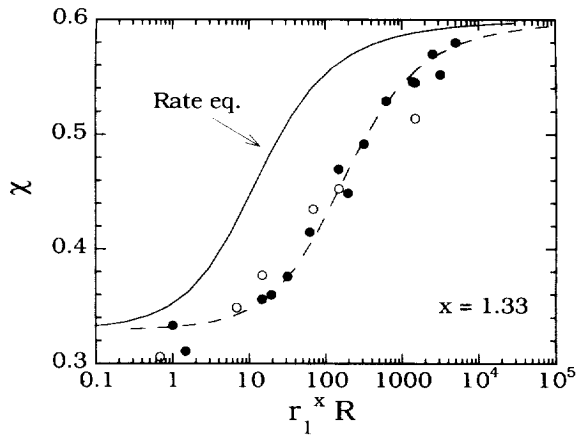

Fig. 3. Scaling exponent $\chi = d(\ln N)/d[\ln(F)]$ as a function of $r_1^{1.33} R$ obtained from simulations of the restricted pair-bond model on a square lattice. Filled symbols correspond to results with $D_e = 0$ (no edge diffusion), while open symbols correspond to results with $D_e = D$ (rapid edge diffusion). The solid curve is a rate-equation prediction (see text). The dashed curve is a corrected rate-equation fit using an effective-detachment rate given by $\alpha = 0.14$.

pair-bond model on a square lattice. As for the triangular lattice, simulations were carried out both with and without rapid island relaxation via edge diffusion and over a wide range of values of r_1 and R ($2.5 \times 10^{-6} < r_1 < 10^{-3}$, $10^5 < R < 10^{11}$). In the case of rapid edge diffusion ($D_e = D$), dimer and trimer edge-diffusion were suppressed in order to prevent small cluster mobility. In both cases we find a transition from a regime with $\chi \simeq 1/3$ (corresponding to $i = 1$) to a regime with $\chi \simeq 0.6$ (corresponding to a critical island size equal to 3).

The inset of Fig. 2 shows the scaling of the island density N as a function of the detachment rate r_1 in the large-u regime corresponding to $i = 3$ ($\chi \simeq 0.6$). As can be seen, the measured value of the exponent ϕ_3 ($\phi_3 \simeq 0.34$) does not agree with the standard rate-equation prediction $\phi_3 = 0.4$. Taking into account the value of χ ($\chi \simeq 0.58$), this implies a value for the crossover scaling exponent x_{13} ($x_{13} \simeq 1.33$) which is significantly below the expected value of 3/2 predicted by Eqs. (2a) and (2b). Similar results obtained with rapid edge diffusion (and for large u) yield even smaller estimates for ϕ_3 ($\phi_3 \simeq 0.27$–0.29). Thus, while the rate-equation predictions $\phi_3 = 0.4$ and $x_{13} = 3/2$ may

hold in the limit of very small r_1 and very large R, over the range of our simulations the crossover exponents are significantly smaller. It should be noted that this parameter range is well within the typical parameter range observed in experiments.

Fig. 2 also shows the island-density crossover scaling function $f_{13}(r_1^{x_{13}}R)=NR^{1/3}$ (for the case of no edge diffusion) using the value of the crossover scaling exponent obtained from our estimate of ϕ_3 in the inset ($x_{13}=1.33$). As can be seen, there is reasonably good scaling, while the slope of the crossover scaling function for large u is close to the asymptotic value of $-4/15\simeq-0.266$ and is consistent with the values of χ shown in Fig. 3. In contrast, a similar crossover scaling plot (not shown) using the rate-equation prediction $x_{13}=3/2$ yields a value for the slope of the crossover scaling function $f_{13}(u)$ for large u which is much smaller (-0.19), and which is inconsistent with the value of χ observed in this regime. We note that preliminary data showing the deviation of the detachment exponent ϕ_3 from the rate-equation prediction was already noted in Ref. [25], while preliminary results for the crossover scaling function f_{13} were shown in Ref. [27]. Simulation results using a different crossover scaling variable have also been presented in Ref. [26].

Fig. 3 shows our results for the effective scaling exponent $\chi=\mathrm{d}(\ln N)/\mathrm{d}(\ln F)$ as a function of the crossover scaling variable $u=r_1^{x_{13}}R$ with $x_{13}=1.33$. Surprisingly, there is very little difference between the results obtained with rapid edge diffusion (open symbols) and without edge diffusion (filled symbols). This is in contrast to our results on the triangular lattice, and may be due to the fact that for the square lattice, the existence of rapid edge diffusion does not guarantee the stability of all clusters larger than 4. Also shown in Fig. 3 (solid curve) is an estimate for χ which was obtained using rate equations appropriate for the case of rapid island relaxation as in Eqs. (4a), (4b) and (4c), in which we have assumed that dimers and trimers may break up via single-bond detachment while all clusters of four or more atoms are stable [31]. We note that this rate-equation result is virtually identical to the "universal crossover" curve predicted in Ref. [28] for the case of fast island restructuring. As can be seen, there is poor agreement between the rate-equation result and our simulation results, even in the case of fast edge diffusion [32]. This implies that, as for the triangular lattice, the standard rate-equation approach is not sufficient to give a quantitative estimate of the scaling behavior in the crossover regime.

Fig. 3 also shows a fit (dashed line) to our simulation results, in which we have replaced the detachment rate r_1' used in the rate equations by an effective detachment rate $r_{1\mathrm{eff}}'=\alpha r_1'$ with $\alpha=0.14$. We caution, however, that this is merely a fit and does not imply that the behavior may be described completely by the use of such an effective-detachment rate.

Finally, we note that we have also tested the Walton relation in this case by calculating the ratio $N_3\,\mathrm{e}^{E_3/k_BT}/N_1^3=r_1^2N_3/N_1^3$ for different values of D/F and r_1 in the $i=3$ regime. Although the fluctuations are quite large due to the small monomer density and critical island density, our results indicate that the Walton relation holds, at least within the accuracy of the simulations. This indicates that the deviations from the scaling behavior predicted by the standard rate equations are not due to the breakdown of the quasi-equilibrium approximation, but are rather due to the effects already discussed, such as the neglect of correlations.

4. Conclusion

The results of extensive kinetic Monte Carlo simulations using a restricted pair-bond model have been presented and compared with rate equations in order to characterize transitions in critical island size in (100) and (111) metal homoepitaxy. In both cases we find significant deviations between the crossover scaling behavior predicted by the standard rate equations and our simulation results. In the case of the transition from $i=1$ to $i=2$ on a triangular lattice (corresponding to metal (111) homoepitaxy), our results for the detachment exponent ϕ_2 and crossover-scaling exponent x_{12} agree with rate-equation predictions. However, the detailed rate-equation predictions for the crossover behavior do not agree with simulations. In addi-

tion, we found significant differences between the cases with and without rapid edge diffusion. For the case of the transition from $i=1$ to $i=3$ on a square lattice (corresponding to metal (100) homoepitaxy), we find that not only are the rate-equation results unable to predict the detailed crossover behavior, but the effective values of the detachment exponent ϕ_3 and the crossover exponent x_{13} deviate from the standard rate-equation predictions.

We note that these results indicate that both in the case of the transition from $i=1$ to $i=2$ (triangular lattice) and for the transition from $i=1$ to $i=3$ (square lattice), the standard rate equations significantly underestimate the transition temperature from $\chi=1/3$ behavior corresponding to $i=1$ to a higher value of the critical island size. For the case of metal (100) homoepitaxy this implies that the critical transition temperatures for the demise of the "$i=1$" regime and the onset of the "$i=3$" regime are significantly higher than predicted in Ref. [28].

Finally, we note that we have been able to partially correct the rate-equation predictions by the use of an effective detachment rate which takes into account the fact that an adatom which detaches from a cluster is more likely to reattach than to attach to a nearby island. However, this approach is somewhat heuristic and a more systematic approach is needed. One possibility is to modify the standard rate-equation approach by the use of self-consistent capture cross-sections [23], along with a self-consistent calculation of the detachment rate entering into the rate equations. Further work will be needed to determine whether this approach will be able to account for the results presented here.

Acknowledgements

This work was supported by National Science Foundation grant DMR-9520842 and by the Office of Naval Research. Part of this work was carried out using the computational facilities of the Cherry L. Emerson Center for Scientific Computation at Emory University.

References

[1] Y.W. Mo, J. Kleiner, M.B. Webb, M.G. Lagally, Phys. Rev. Lett. 66 (1991) 1998.
[2] H.J. Ernst, F. Fabre, J. Lapujoulade, Phys. Rev. B 46 (1992) 1929.
[3] R.Q. Hwang, J. Schroder, C. Gunther, R.J. Behm, Phys. Rev. Lett. 67 (1991) 3279; R.Q. Hwang, R.J. Behm, J. Vac. Sci. Technol. B 10 (1992) 256.
[4] W. Li, G. Vidali, O. Biham, Phys. Rev. B 48 (1993) 8336.
[5] E. Kopatzki, S. Gunther, W. Nichtl-Pecher, R.J. Behm, Surf. Sci. 284 (1993) 154.
[6] G. Rosenfeld, R. Servaty, C. Teichert, B. Poelsema, G. Comsa, Phys. Rev. Lett. 71 (1993) 895.
[7] J.A. Stroscio, D.T. Pierce, R.A. Dragoset, Phys. Rev. Lett. 70 (1993) 3615; J.A. Stroscio, D.T. Pierce, Phys. Rev. B 49 (1994) 8522.
[8] J.-K. Zuo, J.F. Wendelken, Phys. Rev. Lett. 66 (1991) 2227; J.-K. Zuo, J.F. Wendelken, H. Durr, C.-L. Liu, Phys. Rev. Lett. 72 (1994) 3064.
[9] D.D. Chambliss, R.J. Wilson, J. Vac. Sci. Technol. B 9 (1991) 928; D.D. Chambliss, K.E. Johnson, Phys. Rev. B 50 (1994) 5012.
[10] Q. Jiang, G.C. Wang, Surf. Sci. 324 (1995) 357.
[11] F. Tsui, J. Wellman, C. Uher, R. Clarke, Phys. Rev. Lett. 76 (1996) 3164.
[12] D. Walton, J. Chem. Phys. 37 (1962) 2182; D. Walton, T.N. Rhodin, R.W. Rollins, J. Chem. Phys. 38 (1963) 2698.
[13] J.A. Venables, G.D. Spiller, M. Hanbucken, Rep. Prog. Phys. 47 (1984) 399; J.A. Venables, Philos. Mag. 27 (1973) 697; Phys. Rev. B 36 (1987) 4153.
[14] S. Stoyanov, D. Kashchiev, in: E. Kaldis (Ed.), Current Topics in Materials Science, vol. 7, North-Holland, Amsterdam, 1981, pp. 69–141.
[15] J.A. Blackman, A. Wilding, Europhys. Lett. 16 (1991) 115.
[16] J. Villain, A. Pimpinelli, L. Tang, D. Wolf, J. Physique I 2 (1992) 2107; J. Villain, A. Pimpinelli, D. Wolf, Comments Condens. Mater. Phys. 16 (1992) 1.
[17] L.-H. Tang, J. Physique I 3 (1993) 935.
[18] M.C. Bartelt, J.W. Evans, Phys. Rev. B 46 (1992) 12675; Surf. Sci. 298 (1993) 421.
[19] C. Ratsch, A. Zangwill, P. Smilauer, D.D. Vvedensky, Phys. Rev. Lett. 72 (1994) 3194.
[20] J.G. Amar, F. Family, P.M. Lam, Phys. Rev. B 50 (1994) 8781.
[21] G.T. Barkema, O. Biham, M. Breeman, D.O. Boerma, G. Vidali, Surf. Sci. 306 (1994) L569.
[22] Z. Zhang, X. Chen, M.G. Lagally, Phys. Rev. Lett. 73 (1994) 1829.
[23] G.S. Bales, D.C. Chrzan, Phys. Rev. B 50 (1994) 6057.
[24] M. Schroeder, D.E. Wolf, Phys. Rev. Lett. 74 (1995) 2062.
[25] J.G. Amar, F. Family, Phys. Rev. Lett. 74 (1995) 2066.
[26] C. Ratsch, P. Smilauer, A. Zangwill, D.D. Vvedensky, Surf. Sci. 328 (1995) L599.
[27] F. Family, J.G. Amar, Mater. Sci. Eng. B (Solid State Mater.) 30 (1995) 149.

[28] M.C. Bartelt, L.S. Perkins, J.W. Evans, Surf. Sci. 344 (1995) L1193.
[29] J.G. Amar, F. Family, Thin Solid Films 272 (1996) 208.
[30] Z.-P. Shi, Z. Zhang, A.K. Swan, J.F. Wendelken, Phys. Rev. Lett. 76 (1996) 4927.
[31] In this case the detachment rate r_1' used in the rate equations is equal to r_1, since on a square lattice, all three directions for the diffusion of an atom with one bond lead to detachment.
[32] A plot similar to Fig. 3 using the rate-equation prediction $x_{13}=3/2$ also shows poor agreement between the rate-equation prediction for χ and our simulation results, and is roughly equivalent to a shift of the curves and simulation results in Fig. 3 to the left by a factor of 10.