

Rate equations for the growth of Cu islands on Cu(001)

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Received 9 May 1994; accepted for publication 9 September 1994

Abstract

The kinetics of island nucleation and growth during deposition of Cu atoms on Cu(001) is studied using rate equations. The parameters in these equations are obtained using microscopic calculations of the energy landscape on the surface, previously used in Monte Carlo (MC) simulations. This allows a quantitative comparison between the rate equations and the MC results. Our rate equations take into account atoms that fall on the bare substrate as well as on top of existing islands, the mobility of single atoms and small islands, the coalescence of adjacent islands and the possible separation of atoms from island edges. The rate equations are used to explore the island size distribution and island density as a function of the coverage and deposition rates. These rate equations provide a useful and flexible tool that allows to easily modify particular microscopic properties of the system such as the mobility of small islands or the rate of coalescence and examine their effect while leaving all other features intact.

Keywords: Copper; Growth; Low index single crystal surfaces; Models of non-equilibrium phenomena; Models of surface kinetics; Single crystal epitaxy

1. Introduction

Recent experimental observations of island growth in metallic monolayers provided significant quantitative information about nucleation and growth rates as well as island shapes. In these experiments atoms are deposited on a metallic substrate using molecular beam epitaxy (MBE) at a variety of substrate temperatures and deposition rates. The properties of the resulting surfaces are then examined using atom beam diffraction (ABS), low energy electron diffraction (LEED) and scanning tunneling microscopy (STM).

Island nucleation and growth were observed for Ag on Si(111), Au on Ru(0001), Cu on Cu(001), Pb on Cu(001) and Ni on Ni(001) [1–5]. It was found that in a range of temperatures, in which the adatoms are mobile on the surface, islands start to form at a very low coverage. A variety of growth rate exponents have been measured [1,4] and used to deduce the diffusion coefficient [3]. It was also found that the island shapes are system dependent and can be either compact [5] or fractal-like [2]. The morphology in the submonolayer regime has significant effect on the growth mode of the resulting crystal [6].

Theoretical studies of island growth have been done using both rate equations and lattice gas models [7–17]. The rate equations are coupled differential equa-

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tions that describe the density of islands as a function of time. In a simple approach one considers islands of all sizes as identical and includes only two equations: one for the density of islands and the other for the single atom density. This approach provides information about the scaling of the island density as a function of coverage and deposition rate [12–14]. A more complete approach takes into account the density of islands of each size separately, giving rise to a set of infinitely many coupled differential equations [7–11]. These equations take into account the important microscopic processes in the system such as deposition of new atoms, nucleation of islands, atoms captured by islands and coalescence. This mean field type approach can provide general information on island density and island size distribution but does not include island shapes and spatial correlations [18].

Lattice gas models are studied using Monte Carlo (MC) simulations in which atoms are randomly deposited on a lattice and then hop between nearest-neighbor sites [13–17]. The hopping rate depends on the substrate temperature and on the energy barrier, which varies according to the local environment of the hopping atom. Typically, moves that involve bond breaking have a high energy barrier and are thus less frequent. As a result, atoms tend to cluster and form islands, that grow as more atoms join. The shapes of these islands depend on the spectrum of hopping energy barriers, the symmetry of the substrate and the temperature. They can be fractal-like as in diffusion limited aggregation (DLA) type models [14,17], or compact [15,16]. In DLA type models only single atoms are mobile while an atom that hits a site adjacent to another atom or an island becomes immobile. This model gives rise to islands with fractal-like shapes [14,17]. In the bond breaking model, which is a generalization of the DLA model, the hopping energy barrier is proportional to the number of broken nearest-neighbor bonds. It exhibits fractal-like islands when the bond breaking energy is high and compact islands when it is low [16].

In order to obtain a model that properly describes a given system and allows quantitative comparison with experiments one needs to use the actual hopping rates on the surface in the large number of possible local environments. Recently, the present authors and collaborators studied the island growth during deposition of Cu on Cu(001) using detailed microscopic

information and realistic temperatures and deposition rates [15]. The hopping energy barriers were calculated for the 1024 possible local environments in the 3×4 rectangle around the initial and final sites of the hopping atom, using the atom-embedding method of Finnis and Sinclair [19,20]. We then performed dynamical Monte Carlo simulations of Cu deposition on Cu(001) and explored the island density versus coverage and the island size distribution [21]. We found that in the case of Cu on Cu(001) atoms have high mobility along island edges and as a result island shapes are compact.

In this paper we study a set of rate equations which is based on detailed microscopic properties, and provides quantitative comparisons with the Monte Carlo results. We use the same set of energy barriers used in Ref. [15] for Cu on Cu(001) and compare the MC and rate equation results. The rate equations are introduced in Section 2. Numerical results and comparison to MC simulations are presented in Section 3 and a short summary in Section 4.

2. The rate equations

We study the growth of Cu islands on a Cu(001) surface during deposition at a constant rate r and substrate temperature T . In the MC simulations we typically use a 200×200 square lattice on which the Cu atoms are deposited randomly and then hop between nearest-neighbor sites. The hopping rate is given by

$$h = \nu \exp(-E_B/k_B T), \quad (1)$$

where $\nu = 10^{12} \text{ s}^{-1}$ is the vibration frequency and E_B is the energy barrier for the given move. The barrier E_B can take each value in the list E_B^i , $i = 1, \dots, 1024$, for the 1024 possible local environments, determined by the occupancy of the 10 sites adjacent to the initial and final sites of the hopping atom [15]. When two atoms hop into adjacent sites they form a dimer. An atom which runs into an island joins the island which then grows. We find that some small islands are mobile and thus can run into each other and merge or join larger islands. We also observe that a single atom can separate from an island, although the activation energy for this process is much higher than for hopping of an isolated atom. As the islands grow the fraction of the deposited atoms which fall on top of existing islands

increases. These atoms hop on top of the island until they fall down and join that island. In the regime that we study in this work three-dimensional growth does not occur as discussed later.

In the rate equations we use all the microscopic information used in Ref. [15] and the insight we gained from the MC simulations. The rate equations include four terms: a deposition term that describes the atoms falling on the substrate (or on top of existing adatoms and islands); a diffusion term that describes single atoms or mobile islands that run into other adatoms or islands; a coalescence term that describes the merging of two islands into one larger island; and a separation term that describes atoms that separate from island edges and remain on the surface [22].

2.1. Deposition term

Each cluster of two or more atoms is considered an island, and the island size i is given by the number of atoms in it. In general, islands of size i can take a number of different shapes, however, in the rate equation analysis we consider them as identical. We define N_1 as the density of single atoms and N_i , $i \geq 2$ as the density of islands of size i . These are densities per site of the substrate and therefore the coverage is given by

$$\Theta = \sum_{i=1}^{\infty} i N_i. \quad (2)$$

The atoms are deposited on the surface at deposition rate of r monolayers (ML)/s. The probability that an adatom falls on the bare substrate is proportional to $1 - \Theta$. The probability that it falls on top of an island of size i is proportional to $i N_i$ [23]. From the analysis of the energy barriers in the vicinity of a step we find that atoms that fall on top of an island tend to hop on top of the island until they hit the edge and fall down to the first layer without leaving the island. The formation of a second layer is very unlikely in the temperatures and deposition rates studied here until the first layer is almost complete [24]. In fact, the domain of attraction of an island includes not only its area i but also a strip of sites adjacent to the island edge. Assuming that the island shapes are nearly rectangular with small aspect ratios, the size of this domain of attraction is approximately $C(i) = (\sqrt{i} + 2)^2 = i + S(i)$. Here $S(i) = 4(\sqrt{i} + 1)$ is the area of the strip of sites

adjacent to the island. Atoms that fall inside $C(i)$ join the island upon hitting the surface. This feature is especially important for small islands. The contribution of deposited atoms to the island size distribution is thus given by

$$\begin{aligned} \frac{dN_1^{\text{dep}}}{dt} &= r \left[1 - C(1)N_1 - \sum_{i=1}^{\infty} C(i)N_i \right] \\ \frac{dN_i^{\text{dep}}}{dt} &= r [C(i-1)N_{i-1} - C(i)N_i], \\ i &= 2, \dots, \infty. \end{aligned} \quad (3)$$

If an atom falls on a bare substrate it adds into the single atom density (first equation) while if it joins an island of size i it turns the island into size $i + 1$.

2.2. Diffusion term

The main driving force for nucleation and growth of islands is the diffusion term. Single atoms as well as some small islands are mobile and tend to run into each other and thus nucleate new islands or aggregate into existing ones. The diffusion constant of an island with m atoms is given by D_m . The probability that it will run into an immobile island of size j is proportional to $\sigma D_m N_m N_j$ where σ is the capture coefficient [7]. Therefore, the contribution of diffusion to the island size distribution is given by

$$\begin{aligned} \frac{dN_i^{\text{dif}}}{dt} &= \sigma \sum_{m=1}^{i-1} D_m N_m N_{i-m} \\ &\quad - \sigma \sum_{m=1}^{\infty} (D_i + D_m) N_i N_m, \\ i &= 1, \dots, \infty. \end{aligned} \quad (4)$$

We have calculated the diffusion constants for islands of various sizes based on energy barriers obtained earlier. For the single atoms the hopping barrier is $E_B = 0.7$ eV and its diffusion coefficient is $D_1 = \nu \exp(-E_B/k_B T)$, where $\nu = 10^{12} \text{ s}^{-1}$ is the vibration frequency.

Using the set of energy barriers we explored the mobility of small islands. We found that islands of sizes 2, 3, 5 and 7 are mobile while the mobility of all other islands is orders of magnitude lower. The motion of islands is composed of a sequence of moves. For exam-

ple, in a pair of atoms, one of them first moves into a next-nearest-neighbor configuration. Then they move back into a nearest-neighbor configuration. We found that a somewhat more complex sequence of moves makes islands of sizes 3, 5 and 7 mobile as well. The energy barriers for these moves are all very similar: $E_B = 0.72$ eV for the dimer and trimer and 0.71 for the islands of sizes 5 and 7. For simplicity we will use $E_B = 0.7$ for all mobile islands. We calculated the diffusion constant for the mobile islands and found that $D_2 = 0.5D_1$, $D_3 = 0.25D_1$, $D_5 = 0.04D_1$ and $D_7 = 0.02D_1$. The mobility of all other islands is orders of magnitude lower and thus we use $D_i = 0$. This is because they form compact shapes in which diffusion involves at least one move with an energy barrier larger than 1 eV which makes diffusion negligible.

The capture coefficient σ represents the cross section of a mobile atom or island to hit other atoms (or islands). It has been studied extensively in the literature [7–9,11]. Previous studies indicate that σ varies slowly with island size and the coverage [7], while a recent study based on a comparison between the rate equations and a diffusion equation indicates that in some cases σ may exhibit strong dependence on the island size and the coverage [11]. Our numerical studies indicate that for the case of compact islands and for the coverage range studied in this paper σ shows weak dependence on the island size and coverage. We thus use the approximation in which σ is a constant which is adjusted to the value $\sigma = 7.8$ using the MC data for the island density and island size distribution.

2.3. Coalescence term

As the coverage increases islands start growing into each other and merge. The rate at which islands of size m coalesce with islands of size n is $(G_m + G_n)N_mN_n(S(m) + S(n))$ where G_m is the rate of growth of an island of size m and $S(m)$ is the area of the strip of adjacent sites around the island. This growth rate is given by $G_m = rC(m) + \sum iD_iN_i$, where the first term accounts for atoms falling on top of the island and the second term for single atoms and mobile islands that aggregate into it. Since the contribution of mobile islands to coalescence is negligible compared to single atoms we use a simpler expression of the form $G_m = rC(m) + D_1N_1$. We thus find that the contribution of coalescence to the

island size distribution is

$$\frac{dN_i^{\text{cls}}}{dt} = \sum_{m=1}^{i-1} G_m N_m N_{i-m-1} - \sum_{m=1}^{\infty} (G_i + G_m) N_i N_m. \quad (5)$$

In our simulations we observed that including the term D_1N_1 in G_m causes much coalescence in the very early stages of growth, which is not in agreement with the MC data. This is due to the fact that the rate equations do not include spatial correlations between the islands and the fact that at low coverage the island separation is much larger than the island size. At higher coverage the single atom density decreases and then the $rC(m)$ term becomes dominant in G_m . We find that the best fit to the MC data is obtained when $G_m = rC(m)$ and the second term is ignored. Our guess is that this is due to the fact that at higher coverage, when coalescence is important, the single atom density is already low. Also, atoms hopping on the substrate are unlikely to reach the narrow gaps between islands where they can cause coalescence. To prevent the creation of a finite density of extremely large islands, due to repetitive coalescence, we do not allow the creation of islands more than 2.5 times larger than the average island size by coalescence. We believe that this is a reasonable procedure for low coverages, although not for coverages approaching the percolation threshold.

2.4. Separation term

Atoms on the edge of an island may in principle hop away and separate from the island. Since for Cu on Cu(001) islands form compact shapes the number of typical configurations on the edge is small. For a corner atom, the barrier to hop to a neighboring site is 1.1 eV. As the backward move has an energy barrier of 0.3 eV, and the move further away from the island an energy barrier of 0.7 eV, the total barrier for a corner atom to leave an island is $E^{\text{sep}} = 1.1 - 0.3 + 0.7$ eV = 1.5 eV which makes this effect completely negligible for the substrate temperatures we study here. Some islands have an atom on the edge which has only one nearest neighbor. This is the case for islands of sizes 2, 3, 5 and 7 but may also occur for larger islands. For such edge atoms, the barrier for moving into a next-

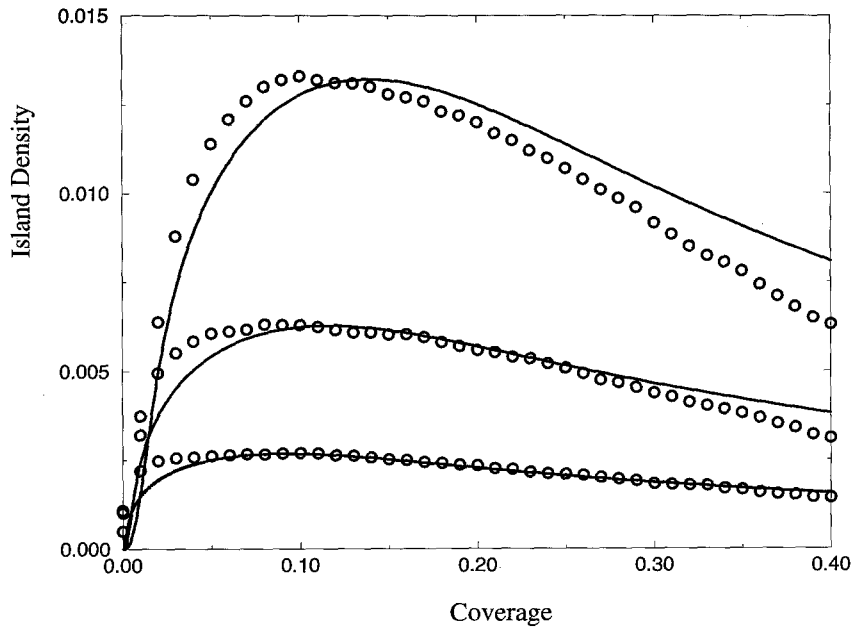


Fig. 1. The island density N versus coverage θ obtained from the rate equations at $T = 350$ K and the deposition rates (from top to bottom) $r = 1$ ML/10 s, 1 ML/100 s and 1 ML/1000 s. The rate equation results (solid line) are in reasonable agreement with the MC results (circles).

nearest-neighbor site of a corner atom is 0.7 eV. As the backward move is 0.3 eV and the move further away from the island is 0.7 eV, the total activation energy for separation is $E^{\text{sep}} = 1.1$ eV.

The contribution of the process of atom separation from islands to the island size distribution is given by

$$\frac{dN_1^{\text{sep}}}{dt} = B_2 N_2 + \sum_{m=2}^{\infty} B_m N_m, \quad (6)$$

$$\frac{dN_i^{\text{sep}}}{dt} = B_{i+1} N_{i+1} - B_i N_i,$$

where $B_i = \nu C_i \exp(-E_i^{\text{sep}}/K_B T)$, is the atom separation rate from islands of size i , E_i^{sep} is the separation energy barrier for the most easily separable atoms in the island and C_i is the number of such atoms in the island. Each atom that separates from an island adds to the single atom pool (first equation), while the island becomes smaller. Since the lowest energy barrier for an atom to separate from an island is higher than 1 eV this process is negligible for the temperatures we study here. At higher temperatures and longer time scales it should become important.

3. Simulations and results

Since all processes mentioned above occur simultaneously the complete set of rate equations is:

$$\frac{dN_i}{dt} = \frac{dN_i^{\text{dep}}}{dt} + \frac{dN_i^{\text{dif}}}{dt} + \frac{dN_i^{\text{cls}}}{dt} + \frac{dN_i^{\text{sep}}}{dt}, \quad i = 1, \dots, \infty. \quad (7)$$

We have simulated Eq. (7) for deposition rates between 1 ML/10 s to 1 ML/1000 s and a temperature of 350 K. To integrate these differential equations we use a fourth order Runge–Kutta routine, typically with a time step of $h = 0.1$ s. Since Eq. (7) may include an infinite set of equations we start with a small number of equations (typically 50 equations for $i = 1, \dots, i_{\text{max}} = 50$) and then increase i_{max} whenever for any $i > i_{\text{max}}$ $dN_i/dt > 10^{-6}r$.

We examined the island size distribution as a function of the coverage for different deposition rates and compared to MC results obtained for the same parameters. To simulate the island growth using rate equa-

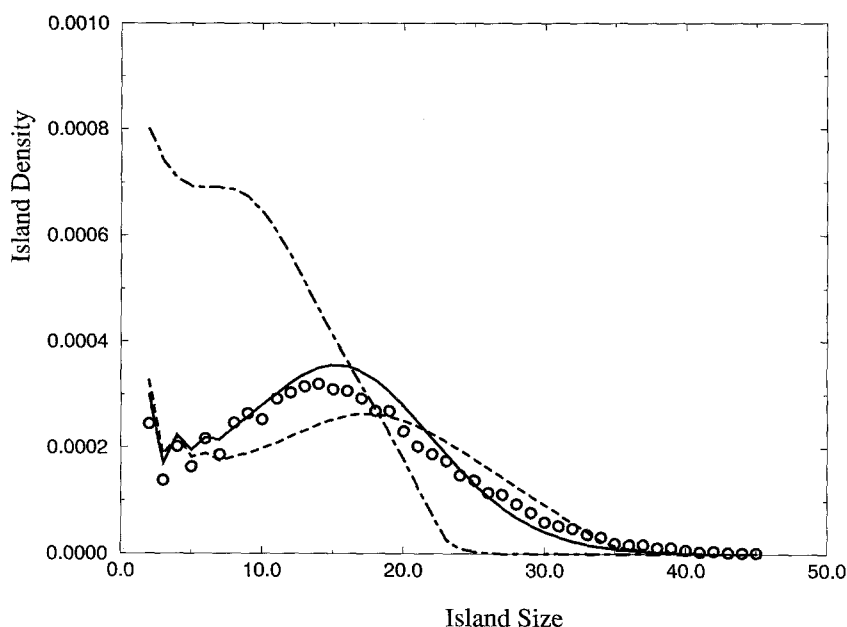


Fig. 2. The island size distribution for $T = 350$ K, deposition rate of $r = 1$ ML/100 s, and $\Theta = 0.1$. The rate equation results (dashed line) are shifted to the right compared with the MC results (circles). A better fit is obtained for rate equations in which the coalescence term is suppressed (solid line). Note that the densities of islands of sizes 3, 5 and 7 are relatively low due to their mobility causing oscillations. Finally, rate equations in which the small-island mobility is suppressed give a completely different distribution (dashed-dotted line) in which the peak is suppressed. Note that in this case the peak simply starts to appear at a somewhat higher coverage.

tions we start with a clean substrate ($N_i = 0$ for all i) and then simulate Eq. (7) up to the desirable coverage $\Theta < 1$. We believe that our rate equations provide a good description at least up to $\Theta = 0.4$.

Considering all clusters of two or more atoms as islands the island density is given by $N = \sum_{i=2}^{\infty} N_i$. In Fig. 1 we plot the island density as a function of the coverage for deposition rates of $r = 1$ ML/10 s, 1 ML/100 s and 1 ML/1000 s at $T = 350$ K. We observe that the rate equations (solid lines) are in reasonable agreement with the MC data (circles). We observe that the island density quickly increases and then saturates. For low deposition rates it remains approximately a constant for a while and then decreases due to coalescence.

In Fig. 2 the island size distribution is shown for $T = 350$ K, $r = 1$ ML/100 s and $\Theta = 0.1$. We find that the island size distribution has a well defined peak. Due to the mobility of islands of sizes 3, 5 and 7 their densities are relatively low causing oscillations in the distribution between sizes 2 and 8. The peak for the

rate equations (dashed line) appears to be shifted to the right compared to the MC data (circles). A better fit is obtained when the coalescence term is suppressed at this low coverage (solid line). We believe that since the rate equations do not include spatial correlations they overestimate the coalescence at low coverage. When the mobility of small islands is suppressed a completely different distribution is obtained (dashed-dotted line). Due to the suppression of their mobility the density of these small islands is much higher and the peak forms only at higher coverage. This demonstrates the importance of the small-island mobility.

We find that as the coverage increases, the peak in the island size distribution shifts to the right and becomes broader as the coverage increases. The island size distribution obtained using the rate equations for $T = 350$ K, $r = 1$ ML/100 s and the coverages $\Theta = 0.1$, 0.2 and 0.3 is shown in Fig. 3. Obtaining the island size distribution from MC simulations is extremely time consuming due to the large number of runs which are

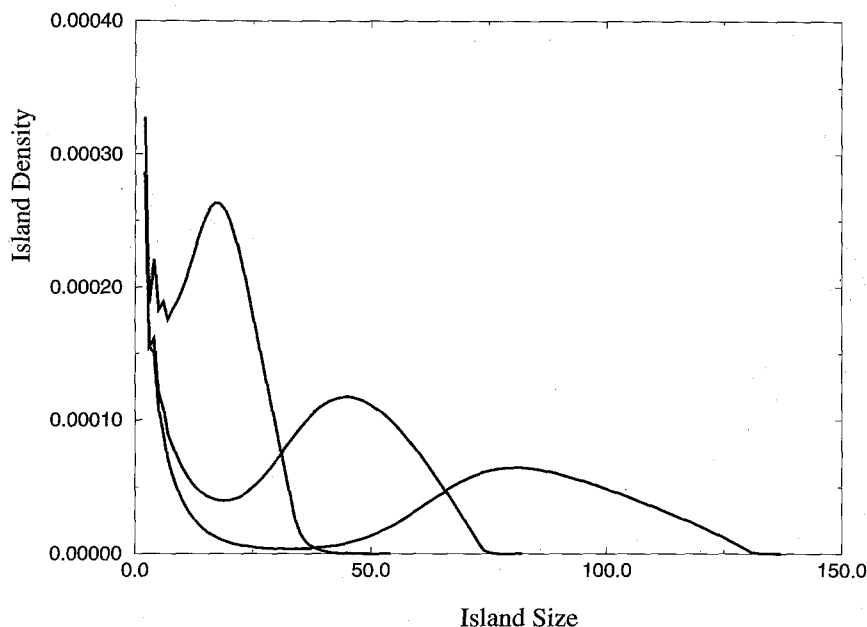


Fig. 3. The island size distribution obtained from the rate equations for $T = 350$ K, $r = 1$ ML/100 s, and $\theta = 0.1, 0.2$ and 0.3 ML. As the coverage increases the peak shifts to the right and becomes broader.

required in order to obtain good statistics. Using rate equations, on the other hand, one needs a single run for each choice of r and T . Therefore, rate equations are especially useful for obtaining more data points, once it is determined that they provide results in agreement with MC.

4. Discussion

We have studied island growth in Cu on Cu(001) using rate equations. We used the insight developed from MC simulations of the same system and realistic parameters which allow direct comparison with the MC results. The rate equations include four terms: the *deposition term* which describes atoms deposited on top of the bare substrate or existing islands, the *diffusion term* which describes nucleation and growth of islands due to diffusion, the *coalescence term* that describes islands that merge and the *separation term* which describes atoms that separate from islands. Our rate equations have been applied for Cu on Cu(001) and include specific properties of this surface such as mobility of small islands. However, we believe that

the procedure used here should apply to other materials once the necessary microscopic information is obtained.

The rate equations provide useful insights into the kinetics of island growth. Since the simulation of rate equations is orders of magnitude faster than an equivalent MC program one can use them for conditions in which MC is not feasible, such as high temperatures or low deposition rates. Due to the simplicity and flexibility of the rate equation approach one can use them to examine the significance of various processes. As an example, to examine the effect of the mobility of small islands (dimers, trimers, etc.) one can easily suppress this mobility and see how it affects the results. Doing a similar manipulation in the MC simulations is extremely complicated, since the mobility is determined by microscopic barriers and any modification would affect many other processes as well.

Acknowledgements

We would like to thank D.O. Boerma, D. Mukamel and G. Vidali for helpful discussions. O.B. thanks the

NSF for support under grant DMR-9217284. G.T.B. would like to acknowledge support by NSF grants ASC-9310244 and DMR-9121654 through the Materials Science Center and the Cornell National Supercomputing Facility and M.B. thanks the NWO for support under the STW program. This research was conducted using the resources of the Cornell Theory Center which receives major funding from the NSF and NYS and additional funding from DARPA, NIH, IBM and other members of the center's Corporate Research Institute.

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- [21] We find that for Cu on Cu(001) the exchange mechanism involves higher energy barriers than hopping and it is thus suppressed. In systems where it appears one can incorporate it into the MC framework. Our approach should also apply for the homoepitaxial growth on the (001) surface of other fcc metals such as Ni, Au, Pt and Ag during deposition.
- [22] An evaporation term, that describes atoms that evaporate from the surface into the gas phase is not included since the energy barrier for this is approximately 3 eV which makes it negligible in the temperature range that we consider.
- [23] We assume that deposited atoms do not show transient mobility. Earlier calculations indicate that for the systems studied in this paper, transient mobility plays a minor role.
- [24] Our barrier calculations show that there are many sites along the island edge where the barrier for falling down is comparable to the ordinary hopping barrier. Therefore, an atom falling on top of an island will hop on top until it will fall down. The smallest immobile island has four atoms and it is very unlikely that four atoms will cluster on top of an island simultaneously. Typically, at low deposition rate, each one of them will fall down before the next one will hit the island. Only at high coverage, when islands are already large the next layer will start to form.