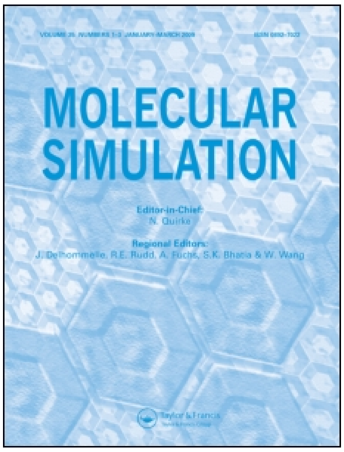


Molecular Simulation
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713644482

An Investigation of Adsorption-Induced Smoothing Mechanisms in Pt/Pt(111) Homoepitaxy

Ruoping Wang[a]; Kristen A. Fichthorn[a]
[a] Department of Chemical Engineering, Pennsylvania State University, PA

To cite this Article Wang, Ruoping and Fichthorn, Kristen A.(1993) 'An Investigation of Adsorption-Induced Smoothing Mechanisms in Pt/Pt(111) Homoepitaxy', Molecular Simulation, 11: 2, 105 — 120
To link to this Article: DOI: 10.1080/08927029308022503
URL: http://dx.doi.org/10.1080/08927029308022503

Molecular Simulation, 1993, Vol. 11(2-4), pp. 105-120
Reprints available directly from the publisher
Photocopying permitted by license only

AN INVESTIGATION OF ADSORPTION-INDUCED SMOOTHING MECHANISMS IN Pt/Pt(111) HOMOEPITAXY

RUOPING WANG and KRISTEN A. FICHTHORN

Department of Chemical Engineering, Pennsylvania State University, University Park, PA 16802

(Received December 1992, accepted January 1992)

We report the results of a molecular dynamics study of the deposition of Pt atoms on a Pt(111) surface. The interatomic potential in our study is generated from Corrected Effective Medium theory. The transient mobility, the interaction between an atom and a ascending step edge, and the incorporation of an atom into a descending step edge have been investigated. No significant transient mobility is observed. Adatoms exchange with atoms at the descending step edges rather than jumping over the edges, and the exchange can involve two or more atoms. Our simulations indicate that the exchange is assisted by instantaneous momentum transfer between the impinging atom and the island atoms.

KEY WORDS: Metal homoepitaxy, molecular dynamics, adsorption dynamics, surface diffusion

1 INTRODUCTION

Recently, there has been much interest in understanding the microscopic mechanisms of thin-film growth [1-18]. Since epitaxy is generally a nonequilibrium process, the atomistic kinetics of deposition and surface diffusion are expected to play an important role in determining film morphology. Atoms randomly deposited onto a substrate are likely to form three-dimensional islands if they stick to where they hit the surface and stack up on each other [7]. In metal homoepitaxy, smooth, layer-by-layer growth is intuitively perceived as resulting from significant thermal mobility of the deposited atoms, which enables them to jump to the energetically most-favored sites [5]. However, recent experiments have demonstrated layer-by-layer growth at very low temperatures for several metal systems [12]. Also, Kunkel *et al.* [13] observed that for Pt/Pt(111) the growth mode is layer-by-layer at high surface temperatures (>621 K), three-dimensional at intermediate temperatures, and, re-entrant into the layer-by-layer mode at low temperatures (< 275 K). These interesting phenomena lie outside of a scenario in which the smoothness of the film is governed by the extent of adatom thermal mobility. Although the real physics is unclear at the present stage, it has been suggested that novel microscopic phenomena can contribute to the smoothness of thin film growth [1, 2, 5, 6, 7, 9].

Several models have been proposed to explain preferential filling of the lowest incomplete layer - a critical factor for achieving layer-by-layer growth - at temperatures too low for thermally activated diffusion. Basically, these models present the following pictures: (1) Interlayer transport is facilitated by a small barrier for interlayer jumping. The "island size and shape" model [13] suggests that small islands or islands with rough, irregular edges, both due to reduced adatom mobility at low temperatures, have a negligible barrier for adatoms to jump over the edge. (2) Smoothing of the film is enhanced by the high kinetic energies of the deposited atoms. In a typical experimental study of metal homoepitaxy, the deposited atoms originate from a "hot" source (for example, a hot filament [15]) which vaporizes them. An impinging atom must transfer its kinetic energy to the surface in order to equilibrate to the lower surface temperature. In the "knock out" model [5], an impinging atom displaces an atom on the partially filled layer in a "one-to-one" ballistic collision and takes its place. The "transient mobility" model [12] postulates that the kinetic energy dissipation of an impinging atom is inefficient and requires an arbitrary time period. During this time period, the deposited atom can use its latent heat of condensation to move across the surface and come preferentially to rest at growing island edges. The "funneling down" [5] model suggests that an impinging atom falls downward along an island edge until it reaches an adsorption site at the bottom of the island, which has the greatest "capture zone". In this way, adsorption sites at the edge of an island in the lowest unfilled layer of the film have higher adsorption rates than sites in higher unfilled layers.

While it is difficult to directly verify these hypotheses with experiments, molecular simulation is an ideal probe which presumably reproduces real microscopic processes and tests the validity of theoretical models with "computer experiments". In this paper we discuss the results of a molecular dynamics simulation study of the deposition of Pt atoms onto a Pt(111) surface. We focus mainly on atomic motion during a short-time span after a deposited atom strikes the surface. We have studied three issues: (1) the "transient mobility" of Pt on Pt(111), (2) the interaction between an impinging atom and an ascending step edge, and (3) the incorporation of atoms into a descending step edge. Our simulations either directly or indirectly address the models discussed above, and reveal new phenomena which should be considered in the interpretation of experimental results. Section 2 will introduce the simulation techniques which we have used for this work, and Section 3 will present the simulation results and a discussion.

2 METHODOLOGY

In this study we employ a computational method for metal deposition on metal surfaces developed by Depristo and coworkers-SCT89 [19, 20]. This method has been widely applied to study metals and related systems [1, 2, 3, 4, 6] and has demonstrated good agreement with experiments and high computational efficiency [19]. In SCT89, the metal-metal interaction potential is modeled using Corrected Effective Medium (CEM) theory [19, 20]. The central idea of CEM theory is that a real N-atom system can be replaced by N effective systems, each of which is an atom embedded into and interacting with a uniform jellium background. The total energy of the interacting atoms is written as:

$$\Delta E\left\{\sum_i A_i\right\} = \sum_i \Delta E\{A_i, n_i\} + [\Delta V_c + \Delta G], \qquad (1)$$

or,

$$\Delta E\left\{\sum_i A_i\right\} = \sum_i \Delta F\{A_i, n_i\} + \Delta V_c, \tag{2}$$

for further simplification. Here, A_i is the ith atom and n_i is its jellium density. The first term sums up the energy of each atom-in-jellium system, which is given by the embedding function ΔE or ΔF. The embedding function is independent of the structure of the specific system. It is a function only of the jellium density and the identity of the atom embedded in it. Embedding functions are available for many elements [19, 21, 22]. The second and third terms, ΔV_c and ΔG, are corrections which account for the difference between the Coulomb energy, the exchange-correlation energy, the kinetic energy, etc. of the real system and the atom-in-jellium system.

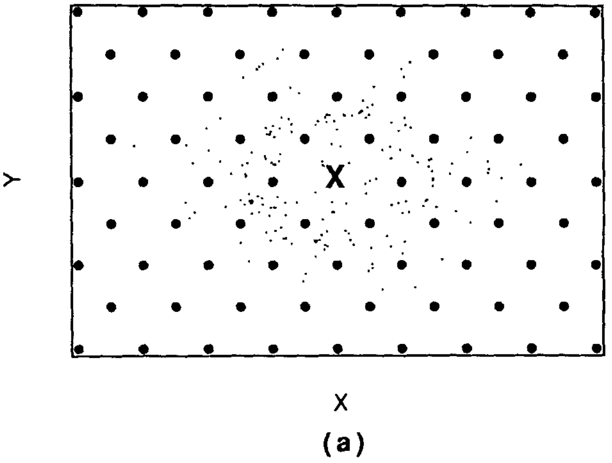
The system is composed of surface atoms, adsorbate atoms, and one gas atom. The surface is modeled by a slab of seven (111) layers, each of which is a square having 18 × 18 surface unit cells. No periodic boundary conditions are applied to the surface. Atoms within a 10 × 10 square in the center of the top three layers are allowed to move, while the rest of the atoms, which represent the infinitely extended system, are fixed. Moving atoms are divided into two classes: inner-active and edge-active [20]. The inner-active atoms interact with the gas atom directly, and they follow Newton's equation of motion. Edge-active atoms are defined as those having at least one fixed atom as their neighbor. The edge-active atoms follow Langevin dynamics [23] and they simulate heat transfer between the active zone and an infinite, isothermal bulk. The adsorbate atoms and the gas atom also follow Newton's equation of motion. The active zone is updated periodically to follow the position of the gas atom, if necessary.

The dynamical equations of motion are integrated using the Verlet algorithm with a simulation time step of 5.0×10^{-15} s. At the beginning of each simulation, the clean or partially filled surface is equilibrated to a constant temperature. Trajectories of the impinging atoms are initiated from above the surface with several incident angles from the surface normal. The initial kinetic energy of the gas atom is 0.25 eV for most of the simulations. This corresponds roughly to temperatures achieved in experimental studies, where the source of the atoms is a hot filament [15]. Atomic motion is visualized by computer animation.

3 RESULTS AND DISCUSSION

a *Transient mobility of impinging atoms on a clean surface*

In this series of simulations, we have deposited an atom onto a clean surface and monitored the outcome of the trajectory. The atom hits the active zone of the surface which, as discussed above, is in the center of the model surface. The aiming points for each trajectory were distributed randomly and uniformly inside one surface unit cell. We performed simulations at surface temperatures of 80 K and 600 K, and with incident angles of 0, 15, 30, 45, and 60 degrees from the surface normal. The simulation time was 4ps for each trajectory. Figure 1 shows the final positions of 200 trajectories, at both the low (Figure 1a) and high (Figure 1b) surface temperature. At 80 K, the impinging atom rests most likely within the second-nearest neighbor of the aiming point. Only about 12% of the trajectories



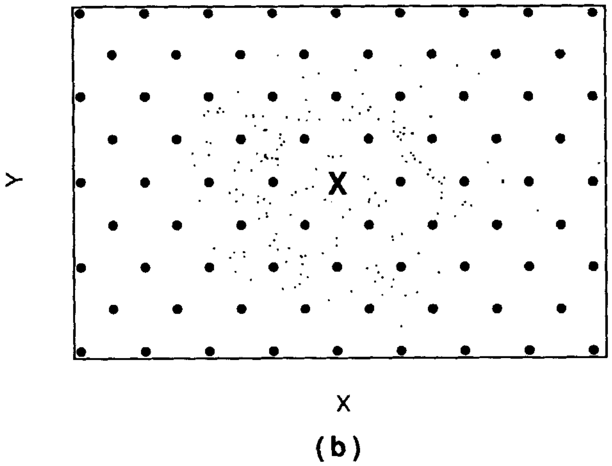


Figure 1 Final positions of Pt atoms deposited onto a clean Pt(111) surface at surface temperatures 80 K (1a) and 600 K (1b). The large circles represent the positions of the Pt surface atoms. Aiming points are inside the unit cell marked with an "X". The simulation time is 4 ps. All trajectories were at normal incidence to the surface.

progressed further than this. At 600 K, the fraction of long jumps increases to about 20%.

Two major factors governing the transient mobility of an impinging atom are the corrugation of the potential-energy surface and the efficiency with which the kinetic energy of the impinging atom is transferred to the surface. To characterize the corrugation of the Pt/Pt(111) potential-energy surface, we have calculated the self-diffusion barrier of a Pt adatom on a clean Pt(111) surface using transition-state theory. As a first approximation, we have kept the surface atoms fixed to their equilibrium positions and considered only the motion of the adatom. Since the

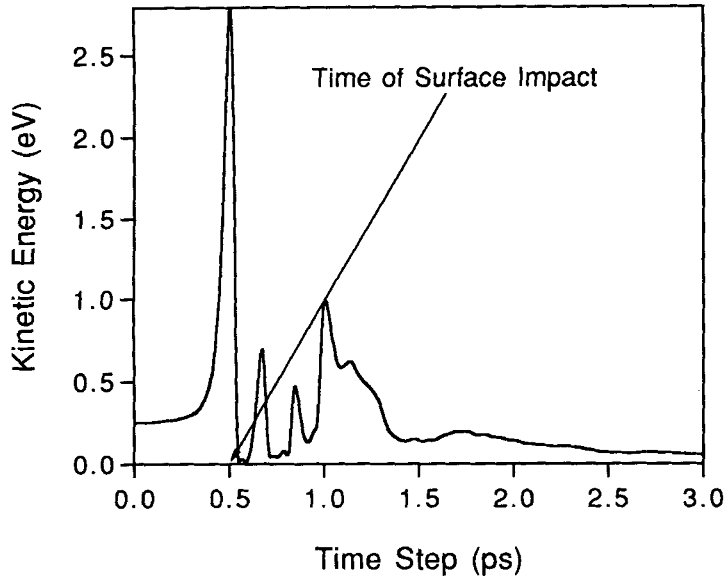


Figure 2 Kinetic energy vs time for of a Pt atom approaching the clean Pt(111) surface. The kinetic energy is an average over 50 trajectories. The surface temperature is 80 K and the initial incident angle is along the surface normal.

fcc(111) surface is closely packed, we do not anticipate the influence of surface relaxation to be significant. Also, Liu *et al.* [8] has found that self-diffusion on Pt(111) does not involve adatom-surface atom exchange. The fcc three-fold site has the lowest energy and the bridge site between two nearest-neighbor Pt atoms is the saddle point on the minimum-potential-energy surface. The diffusion barrier, which we have defined as the energy difference between these two sites, is only 0.18 eV. This value is comparable to the value which Liu *et al.* calculated using an embedded-atom potential [8]. With this low barrier, a small amount of undissipated kinetic energy of the gas atom could make it mobile on the surface. Yet there is no significant transient mobility on this very smooth surface. Our trajectories do exhibit greater mobility than those for fcc metals deposited on the fcc(100) surface [6], on which the diffusion barrier is higher. However, we attribute this to thermal mobility rather than transient mobility, since the Pt/Pt(111) diffusion barrier is small. The increasing percentage of long jumps at 600 K (Figure 1b) indicates the "thermal" nature of adatom mobility. Another piece of corroborating evidence is that the kinetic energy of an impinging atom drops rapidly after its collision with the surface. This can be seen from Figure 2 which depicts an average energy profile of an impinging atom at 80 K. Our results affirm those of the study of Sanders *et al.*, who found no transient mobility in Cu deposition on Cu(111) [2].

b *Interaction between an atom and the ascending step edges*

It is intuitively perceived that an ascending step edge will attract atoms nearby. However, the interaction strength and range and their dependence on the structure

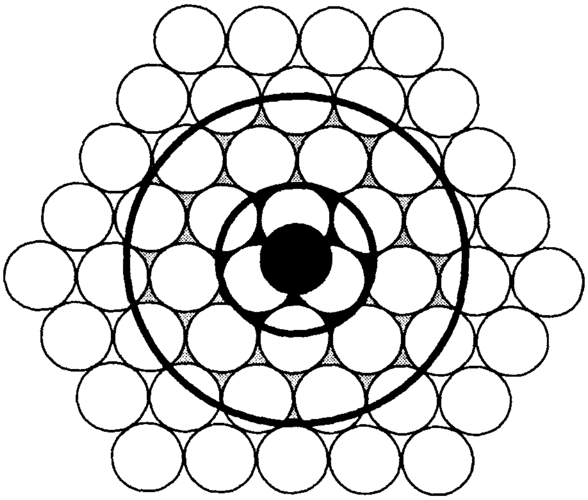


Figure 3 An approximate cross section for the interaction of an impinging atom with an atom adsorbed on an fcc three-fold site, for normal incident angles. The adsorbed atom is represented by the dark filled circle in the center. Atoms impinging within the first concentric region, onto the dark three-fold sites, were repelled from the adatom, while atoms impinging between the first and second concentric rings, onto the gray three-fold sites, were attracted to the adatom. Atoms impinging outside of the second ring were unaffected by the presence of the adatom.

of the step edge involve details of the interatomic forces. In this series of studies, we have examined the possibility that an island could be surrounded by a "capture zone" in which impinging atoms will be drawn to the step edge. If the range of the zone is significant, then atom capture at the step edges could alter the structure of the growing film.

In these simulations we have placed one partially filled layer on top of a completed layer and deposited an atom onto the area outside the island. Beginning with the simplest situation in which the incomplete layer is just one adsorbate atom on an fcc site, we have changed the distance between the aiming point and the island for each simulation run. Our simulations show that the atom can be repelled from, attracted to, or unaffected by the island depending on the distance between them. To characterize the interaction between the impinging and adsorbed atom, we determined an approximate cross section for an atom with normal incidence. A summary of these simulation results is shown in Figure 3. Atoms impinging within the nearest-neighbor fcc site from the adsorbed atom were repelled from it. The repulsion resulted in the ballistic displacement of either the impinging or the adsorbed atom, or both. Trajectories incident upon the intermediate regime in Figure 3 were drawn to the adatom. At distance greater than the second-nearest fcc site or the third-nearest hcp site from the adsorbed atom, we saw no interaction of the two atoms within the 4ps simulation time.

Our trajectory results are consistent with the calculated minimum-potential-energy profile for one atom approaching another atom on an fcc three-fold site, shown in Figure 4. From Figure 4, it can be seen that the attraction between the two atoms is localized to the nearest-neighbor fcc site and that the barrier for

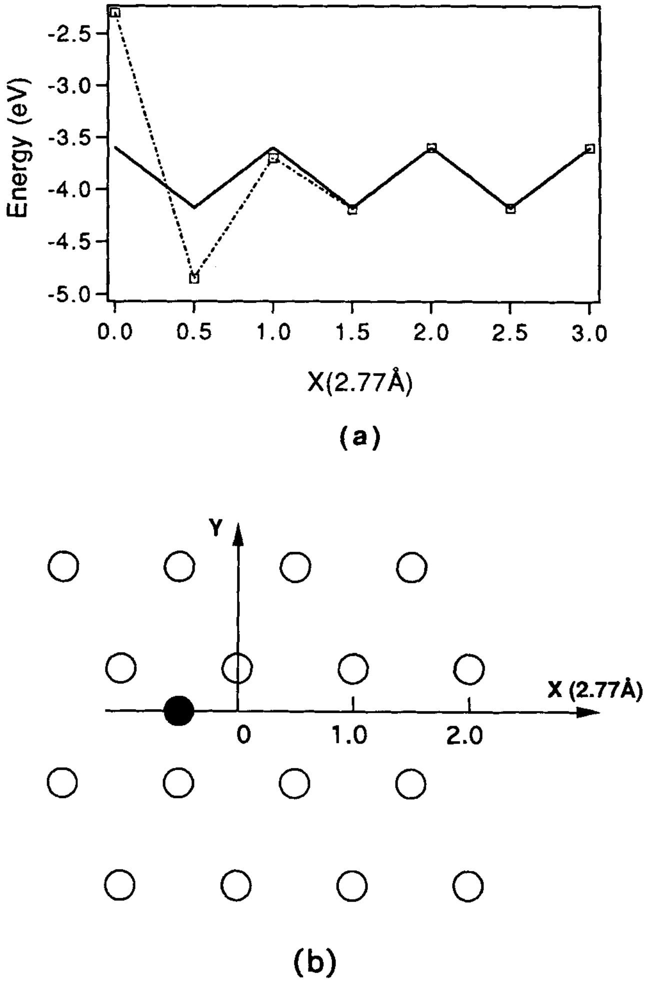


Figure 4 (a) Minimum-potential-energy profiles of an atom on both a clean surface and a surface with one adsorbed atom along the direction indicated by (b). In (b), open circles represent atoms of the completed layer and the solid circle represents the adsorbed atom. The distance along the x-axis is in units of the nearest-neighbor distance of 2.77 Å.

approaching site from the second-nearest-neighbor fcc site is somewhat reduced from the energy barrier for a clean surface. Beyond the second-neighbor distance, the minimum-potential-energy profile is identical to that for a clean surface.

To examine the interaction of an adatom with an irregular step edge, we have constructed two islands, each containing a "kink site" at which an adatom would be strongly bound, and calculated the minimum-potential-energy profile of an atom approaching the kink. Figures 5 and 6 depict the geometry of each kink site and the minimum-potential-energy profile of an approaching atom, respectively. A comparison of Figures 4, 5 and 6 shows that although the binding of the atom to the

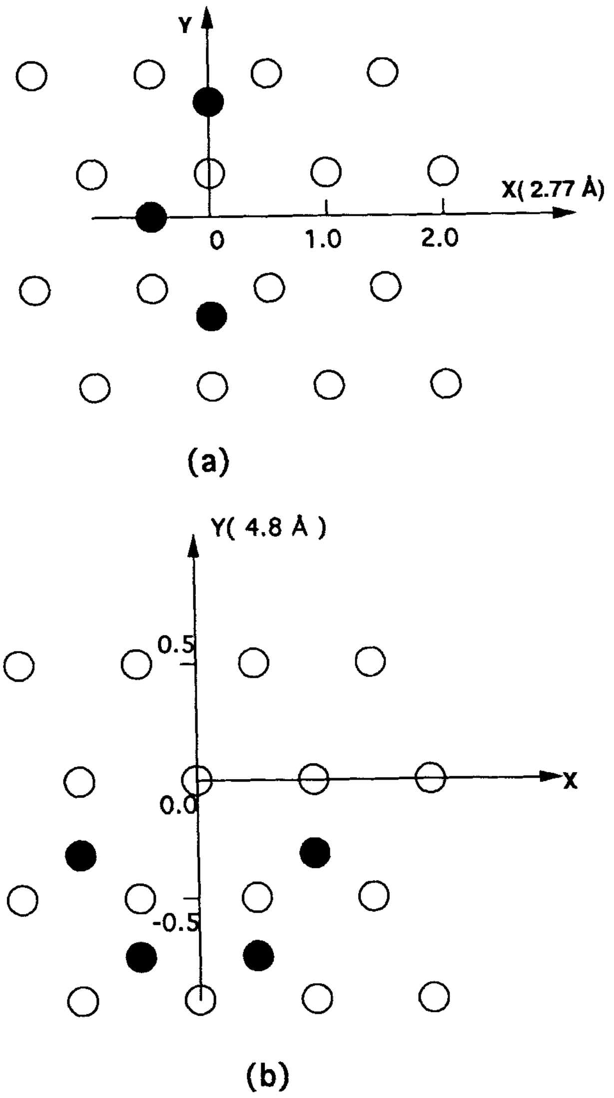
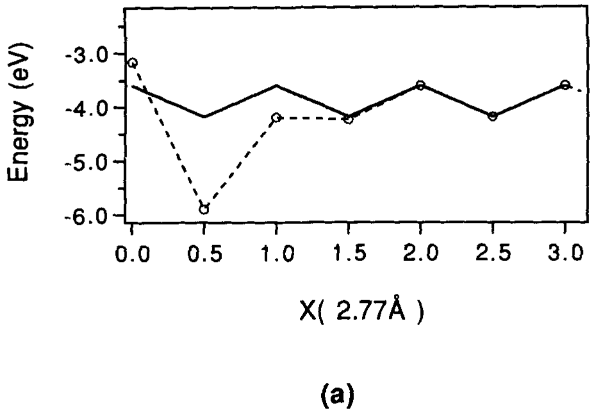


Figure 5 Geometric structures of the ascending step edges we have studied. Open circles represent atoms of the completed layer and solid circles represent island atoms (a) and (b) represent three- and four-atom islands, respectively. In (a) the distance along the x-axis is in units of 2.77 Å, and in (b) the distance along the y-axis is in units of $\sqrt{3} \times 2.77$ Å.

step edge becomes stronger as the number of bonds between the atom and the edge is increased, the range of the interaction is not extended significantly. Hence, the step-edge attraction is localized for the small islands we have studied. Even for larger islands with more edges atoms, the attraction is unlikely to extend out by a great distance. This is because metallic bonding is short-ranged and each adatom basically interacts only with its nearest-neighbor atoms. Adatoms are incorporated into ascending step edges only if they are able to overcome the diffusion barrier



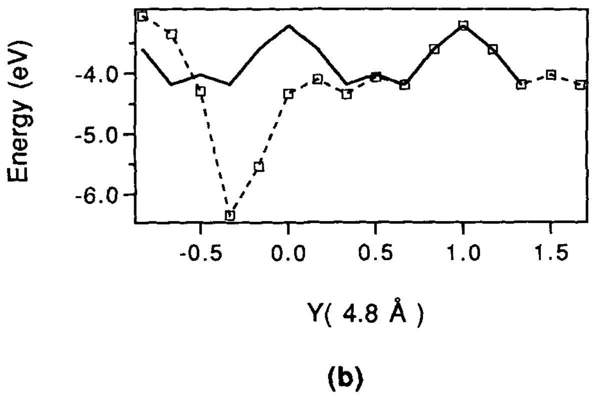


Figure 6 Minimum-potential-energy profiles of an atom on both a clean surface and a surface with the islands shown in Figure 5. The profiles in (a) and (b) are calculated along the lines shown in Figure 5(a) and Figure 5(b), respectively. In (a), the solid line is for a clean surface, and the circles are for the three-atom island shown in Figure 5(a). In (b), the solid line is for a clean surface and the squares are for the four-atom island shown in Figure 5(b).

to approach the step edges. This picture is consistent with the FIM experiments of Wang *et al.* for Ir/Ir(111)[14]. They reported that incorporation of an Ir atom at the ascending step edge of an Ir island occurs once adatom thermal diffusion becomes significant.

c *Incorporation of an atom into descending step edges*

In this series of simulations we have deposited atoms onto the tops of islands consisting of nine and sixteen atoms. The structures of these islands are shown in Figure 7. We will discuss the results of simulations with a nine-atom island in detail, since this system has been examined the most thoroughly. We have changed the surface temperature, the aiming point, the initial incident angle, and the initial kinetic

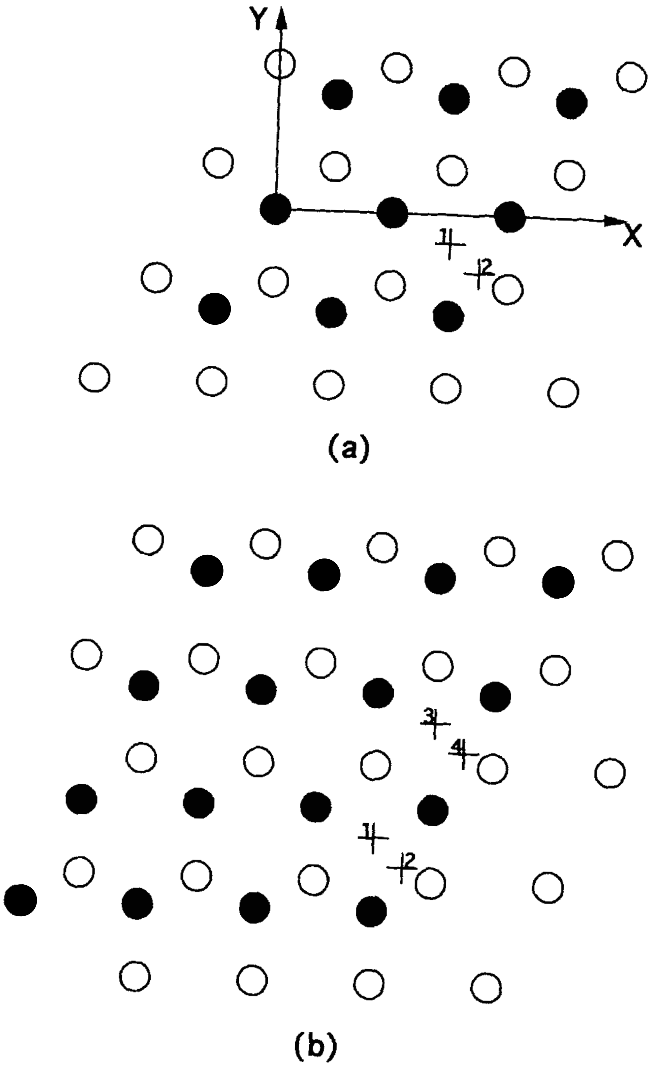


Figure 7 Geometric structures of the descending step edges we have studied and possible pathways for hopping over the step edges. Open circles are atoms of the completed layer and solid circles are island atoms. In (a), $E_1 = -3.08\,\text{eV}$, and $E_2 = -3.48\,\text{eV}$. In (b), $E_1 = -3.85\,\text{eV}$, $E_2 = -3.43\,\text{eV}$, $E_3 = -4.00\,\text{eV}$, and $E_4 = -3.49\,\text{eV}$.

energy, and run 50 trajectories for each set of simulation conditions. The simulation time was 6 ps for each run. Table 1 summarizes our results which present the following interesting features.

First, an atom impinging onto the top of the island descends the step edge by "pushing out" the edge atoms rather than by jumping over the edge. The same process has been observed in simulations with a sixteen-atom island. Sequential snapshots in Figure 8 illustrate the exchange mechanism for the sixteen-atom island.

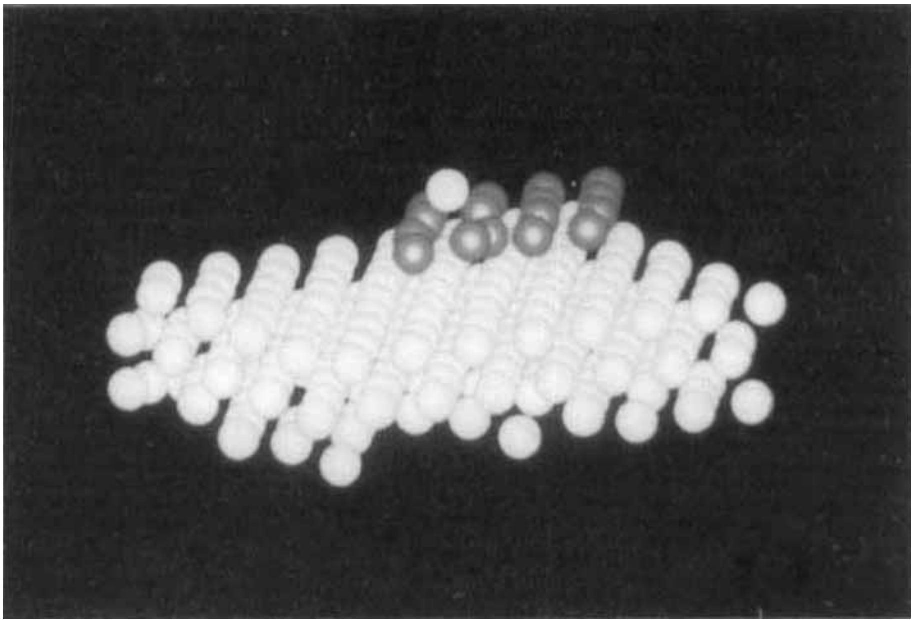


Figure 8a Sequential snapshots illustrating the exchange mechanism for an atom impinging on the edge of a sixteen atom island. The surface temperature is 80 K and the initial incident angle is along the surface normal. The gold balls represent atoms of the completed layers, the red balls represent island atoms, and the blue ball represents the impinging atom. (see colour plates)

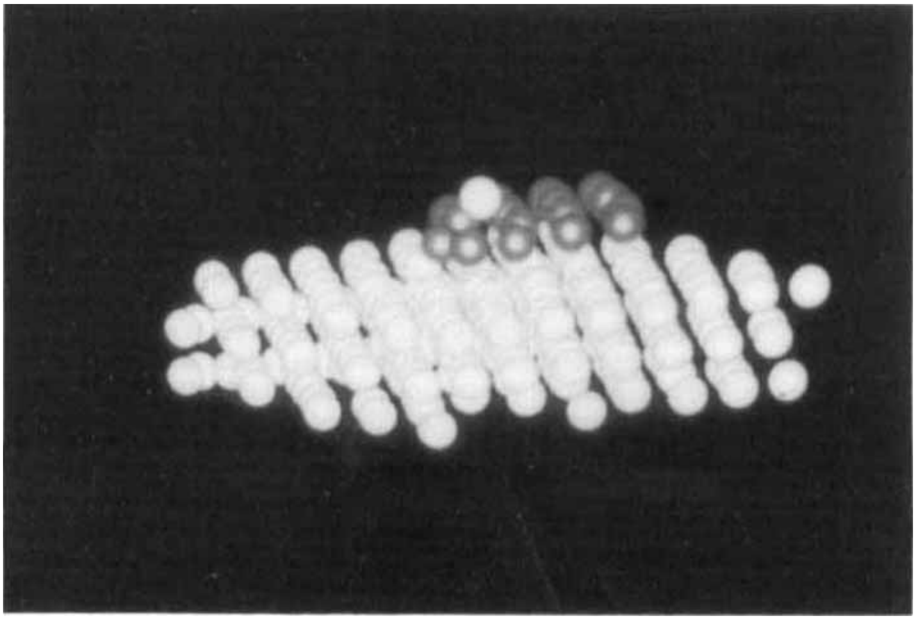


Figure 8b

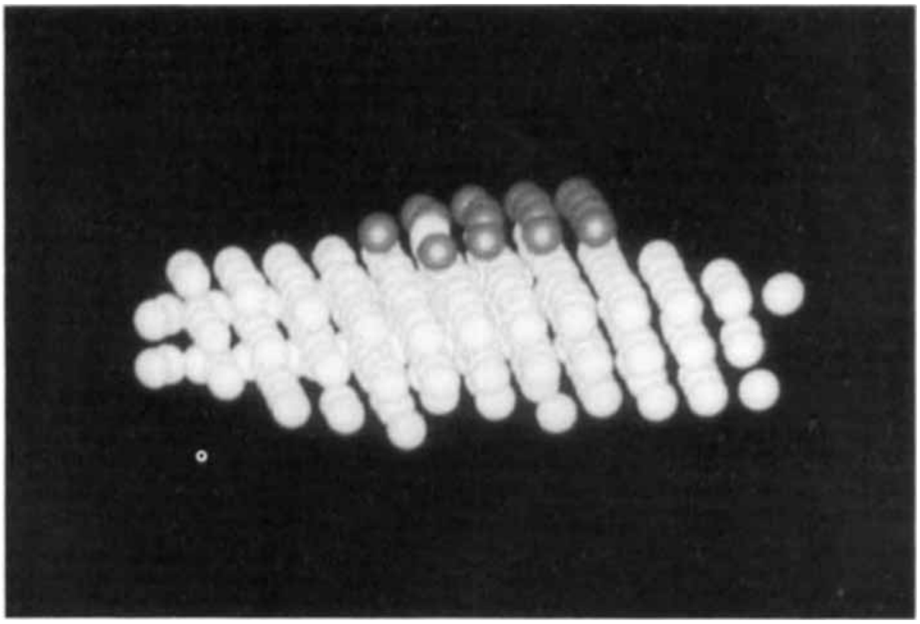


Figure 8c

In most cases, only one edge atom is displaced by the impinging atom. However, we have found exceptions at both high and low temperatures in which an impinging atom displaces several atoms in the island in its descent to the lower level. Figure 9 portrays the rearrangement of atomic positions in two of these instances. Evidence for an exchange mechanism at the descending step has been seen experimentally in FIM studies of the W/Ir(111) system [14]. We are aware that Jonsson has observed the exchange process in a study of the Pt/Pt(111) system using an embedded-atom potential [24].

Although adatom hopping over the step edges might be envisioned as a reasonable channel for interlayer transport, we did not observe this event in any of our simulation runs. To assess the likelihood of hopping over the step edge, we have estimated the barrier for adatom hopping from a three-fold site on top of an island to a three-fold site at the island edge via the bridge site at the island edge. We have considered three possible pathways for the nine- and sixteen-atom islands shown in Figure 7. Again we have utilized rigid islands for these calculations and calculated the barrier as the minimum-energy difference between the indicated bridge and three-fold sites in Figure 7. From our estimate, the barriers are 0.50 eV for path 1 → 2 [Figure 7(a)], 0.42 eV for path 1 → 2 [Figure 7(b)], and 0.51 eV for path 3 → 4 [Figure 7(b)]. These are much higher than the diffusion barrier on a clean surface. A comparison of the barrier for exchange to the barrier for hopping over the step edge will reveal the relative significance of the two mechanisms on a long time scale.

For the short times which we have probed, the exchange mechanism appears to be the dominant mode of interlayer transport. However, from Table 1, which summarizes the results of the deposition runs, we can conclude that the exchange

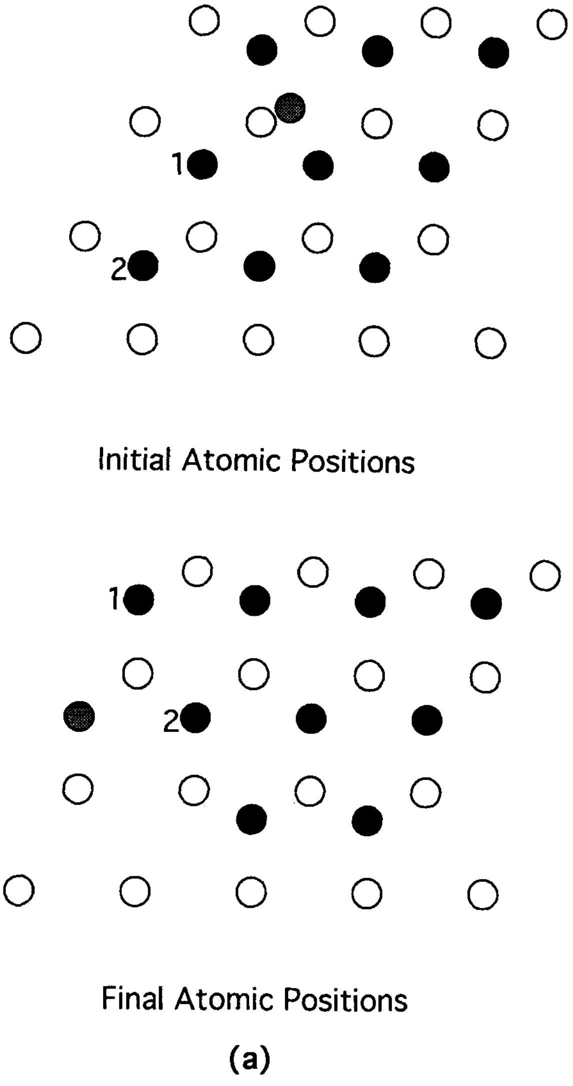
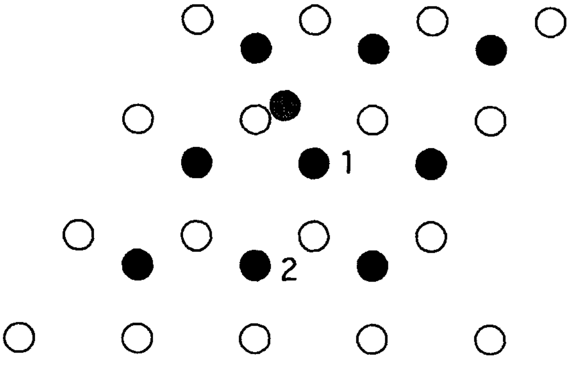
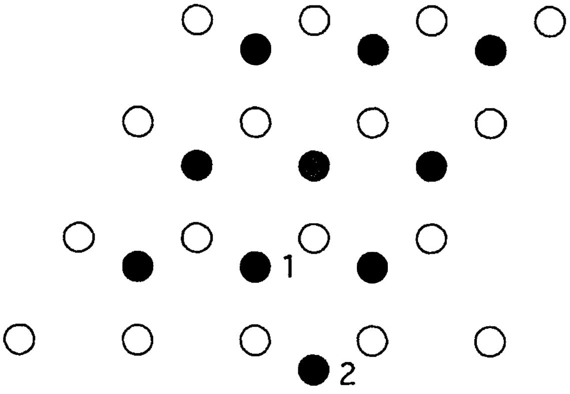


Figure 9a

tendency is significantly enchanced by the momentum of the impinging atom, leaving the relative roles of thermal and non-thermal motion unclear. Comparing Rows 1 and 2 in Table 1, we can see that, at a surface temperature of 80 K, a gas atom with normal incidence hitting a three-fold site at the step edge (referred to as "site 1" in Table 1) pushes out an edge atom, while a gas atom hitting the atom in the center of the island (referred to as "site 2" in Table 1) stays above the three-fold site at the step edge. In the latter case, the momentum of the impinging atom is most likely transferred to the central atom, which is the most tightly bound island atom, while in the former case, the momentum is more easily transferred to the edge atoms. That is why the exchange is difficult (easy) for the later (former) situation. Similarly, comparing Rows 6 and 7 we can see that, at the same surface temperature



Initial Atomic Positions



Final Atomic Positions

(b)

Figure 9 Rearrangements of the island atoms in two types of multi-atom exchange which we observed. Open circles represent atoms of the completed layer, solid circles represent island atoms, and the shaded circle represents the gas atom. Island atoms displaced by the gas atom are numbered. In (a), the exchange involves only step-edge rearrangement, while in (b), an impinging atom displaces both the central atom and an edge atom.

and with the same aiming point, a gas atom with a velocity directed toward the descending step edge (referred to as "(45, 180)" in Table 1) displaces the edge atom, while a gas atom with a velocity directed towards the ascending step edge (referred to as "(45, 0)" in Table 1) most likely stays above. All of these facts indicate that ballistic-momentum transfer assists with the exchange process. Further support for this picture can be seen in the temperature dependence of the exchange. From analyzing Table 1, Rows 1 and 4, we can see that nearly instantaneous exchange is

Table 1 Simulations of the incorporation of an atom into the descending step edge of a nine-atom island.

Total no. of runs	*Stay above*	*Exchange*	*Surface temperature*	*Aiming point*	*Incident angle (Θ, Φ)**	*Initial kinetic energy*
50	0	50	80 K	site 1	(0, 0)	0.25 eV
50	50	0	80 K	site 2	(0, 0)	0.25 eV
50	7	43	80 K	site 1	(0, 0)	0.007 eV
50	6	44	600 K	site 1	(0, 0)	0.25 eV
50	6	44	600 K	site 2	(0, 0)	0.25 eV
50	1	49	80 K	site 1	(45, 180)	0.25 eV
50	42	8	80 K	site 1	(45, 0)	0.25 eV

* Θ is the angle from the surface normal; Φ is the angle from the x-axis in Fig. 7a.

less probable at the higher temperature. Moreover, we found that, at 80 K, most of the exchanges occurred within 2 ps of the collision, while at 600 K, there was a distribution of exchange times. The declining significance of ballistic-momentum transfer in the exchanges at the higher temperature can be attributed to the increasing tendency for random thermal motion of the island atoms, which reduces the probability for a direct, one-to-one collision of an impinging atom with an edge atom.

A final issue concerning the exchange is that it is apparently not strongly dependent on the initial velocity of the impinging atom. This is because an atom approaching the surface is greatly accelerated by its attraction to the surface, as is illustrated in Figure 2. A comparison of Rows 1 and 3 in Table 1 shows that decreasing the incident kinetic energy to about the surface temperature had no significantly different effect on the exchange at 80 K.

4 CONCLUSIONS

Using molecular dynamics simulations we have investigated several adsorption-induced smoothing mechanisms in Pt/Pt(111) homoepitaxy. We do not observe significant transient mobility of a Pt atom impinging on a clean Pt(111) surface. The interaction between an adatom and an ascending step edge is strong and localized. Adatoms at the descending step edges exchange with edge atoms instead of jumping over the edge. Our results indicate that the exchange is enhanced by instantaneous momentum transfer between the impinging gas atom and the island atoms. This is the only contribution of the kinetic energy of the impinging atom to smoothness in thin film growth that we have found. The microscopic details of the deposition of Pt on Pt(111) are very different from those of several metal/metal(100) systems [1].

Acknowledgements

This work was funded by the National Science Foundation under grant CTS-9058013. The computer workstations were provided by a NSF equipment grant. We acknowledge helpful discussions with the DePristo group at Iowa State University, in particular, with Dr. David E. Sanders. We also thank Mr. Prakash Balan for his help with the computer animation.

References

[1] J.W. Evans, D.E. Sanders, P.A. Thiel, and A.E. DePristo, "Low-temperature epitaxial growth of thin metal films", *Phys. Rev. B* **41**, 5410 (1990).

[2] D.E. Sanders, D.M. Halstead, and A.E. DePristo, "Metal/metal homoepitaxy on fcc(111) and fcc(001) surfaces: Deposition and scattering from small islands", *J. Vac. Sci. Technol. A* **10**, 1986 (1992).

[3] D.E. Sanders and A.E. DePristo, "Predicted diffusion rates on fcc(001) metal surfaces for adsorbate/substrate combinations of Ni, Cu, Rh, Pd, Ag, Pt, Au", *Surf. Sci.* **260**, 116 (1992).

[4] D.E. Sanders and A.E. DePristo, "A non-unique relationship between the potential energy surface barrier and the dynamical diffusion barrier: fcc(111) metal surface", *Surf. Sci. Lett.* **264**, L169 (1992).

[5] J.W. Evans, "Factors mediating smoothness in epitaxial thin-film growth", *Phys. Rev. B* **43**, 3897 (1991).

[6] D.E. Sanders and A.E. DePristo, "Metal/metal homo-epitaxy on fcc(001) surfaces: Is there transient mobility of adsorbed atoms", *Surf. Sci.* **254**, 341 (1991).

[7] I.K. Marmorkos and S. Das Sarma, "Atomistic numerical study of molecular-beam-epitaxial growth kinetics", *Phys. Rev. B* **45**, 11262 (1992).

[8] C.L. Liu, J.M. Cohen, J.B. Adams and A.F. Voter, "EAM study of the surface self-diffusion of single adatoms of fcc metals Ni, Cu, Al, Ag, Au, Pd, and Pt", *Surf. Sci.* **253**, 334 (1991).

[9] P.I. Cohen, G.S. Petrich, P.R. Pukite and G.J. Whaley, "Birth-death models of epitaxy I. Diffraction oscillations from low index surfaces", *Surf. Sci.* **216**, 222 (1989).

[10] M. Schneider, A. Rahman, and Ivan K. Schuller, "Role of relaxation in epitaxial growth: a molecular-dynamics study", *Phys. Rev. Lett.* **55**, 604 (1985).

[11] W.M. Plotz, K. Hingerl, and H. Sitter, "Monte Carlo simulation of epitaxial growth", *Phys. Rev. B* **45**, 12122 (1992).

[12] W.F. Egelhoff, Jr., and J. Jacob, "Reflection high-energy electron diffraction (RHEED) oscillations at 77 K", *Phys. Rev. Lett.* **62**, 921 (1989).

[13] R. Kunkel, B. Poelsema, L.K. Verheij, and G. Comsa, "Reentrant layer-by-layer growth during molecular-beam epitaxy of metal-on-metal substrates", *Phys. Rev. Lett.* **65**, 733 (1990).

[14] S.C. Wang and G. Ehrlich, "Atom incorporation at surface clusters: an atomic view", *Phys. Rev. Lett.* **67**, 2509 (1991).

[15] S.C. Wang and G. Ehrlich, "Atom condensation on an atomically smooth surface: Ir, Re, W, and Pd on Ir(111)", *J. Chem. Phys.* **94**, 4071 (1991).

[16] H.A. van der Vegt, H.M. van Pinxteren, M. Lohmeier, and E. Vlieg, "Surfactant-Induced layer-by-layer growth of Ag on Ag(111)", *Phys. Rev. Lett* **68**, 3335 (1992).

[17] T. Michely and G. Comsa, "Generation and nucleation of adatoms during ion bombardment of Pt(111)", *Phys. Rev. B* **44**, 8411 (1991).

[18] D. Gibbs, G. Grubel, D.M. Zehner, D.L. Abernathy, and S.G.J. Mochrie, "Orientational epitaxy of the hexagonally reconstructed Pt(001) surface", *Phys. Rev. Lett.* **67**, 3117 (1991).

[19] T.J. Raeker and A.E. DePristo, "Theory of chemical-bonding based on the atom-homogeneous electron gas system", *Inter. Rev. in Phys. Chem.* **10**, 1 (1991).

[20] D.E. Sanders, Ph.D. Thesis, Iowa State University, (1990).

[21] S.B. Sinnott, M.S. Stave, T.J. Raeker, and A.E. DePristo, "Corrected effectfve-medium study of metal-surface relaxation", *Phys. Rev. B* **44**, 8927 (1991).

[22] K.W. Jacobsen, J.K. Norskov, and M.J. Puska, "Interatomic interactions in effective-medium theory", *Phys. Rev. B* **35**, 7423 (1987).

[23] A.E. DePristo and H. Meitu, "Molecular dynamics simulations of energy flow at a solid surface: New methods using a small number of atoms", *J. Chem. Phys* **90**, 1229 (1989).

[24] H. Jonsson, private communication.