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Atomic exchange processes in sputter deposition of Pt on Pt(111)

Marie Villarba, Hannes Jónsson *

Department of Chemistry, BG-10, University of Washington, Seattle, WA 98195, USA

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Abstract

We have conducted a theoretical study of atomic exchange processes occurring as Pt atoms are deposited on a Pt(111) surface. Using an embedded atom method type interaction potential, we have systematically sampled sites atop a heptamer and at straight A and B type steps to correlate incident atom energy and regions of impact with the probability of 'push-out' events, where deposited atoms displace peripheral island atoms. Push-out events lead to incorporation of the incident atom in the growing layer and, thereby, can contribute to layer-by-layer growth if islands are small enough. We find the probability of push-out events to be large for incident energy typical for sputter deposition, 10 and 20 eV. To some extent the 20 eV incident energy also results in detrimental effects, such as displacement of surface atoms (interface mixing), vacancy formation, and displacement of island atoms to positions atop the growing layer, promoting multilayer growth. We have evaluated the minimum energy paths and activation energy barriers for several representative exchange processes observed in the simulations. The calculated activation energy is surprisingly low, typically in the range of 0.6–0.8 eV, even for processes involving displacements of multiple atoms.

1. Introduction

Thin films are important for a host of devices with unique properties – electronic, superconducting, magnetic, optical, etc. The performance of each product for its intended function is strongly dependent on the orderly arrangement of atoms within the films. A reduction of process temperature in thin film growth would allow improvement in film quality as many undesirable processes, such as interface mixing, are minimized at lower temperatures. For the epitaxial growth of semiconductor films, ion beam sputter deposition (IBSD) seems suited for low temperature growth because of the suprathermal energy ($E_{inc} \approx 10$ eV) of condensing particles and the feasibility of high vacuum growth [1]. To optimize thin film growth using such sputter deposition techniques, a thorough understanding of the atomic-scale processes governing surface morphologies in sputter deposition is required.

We present here results of computer simulation studies of sputter deposition where several atomic scale processes affecting the surface morphology are identified. We are not aware of experimental studies of these effects, but have chosen to simulate Pt deposition on Pt(111) since vapor deposition and sputtering have been studied extensively for that system using both thermal He atom scattering [2] and STM [3]. In our previous simulations of Pt vapor deposition at 275 K [4], we found deposited atoms directed atop an island can use their latent heat of

* Corresponding author.

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condensation to displace peripheral island atoms to become incorporated at the island edge. This 'push-out' effect can, therefore, enhance layer-by-layer growth. Similar effects have been observed in simulations of Cu vapor deposition onto Cu(111) [5]. While the kinetic energy of incoming vapor atoms is very small, 0.25 eV, the atoms gain nearly 4 eV as they approach the surface due to the attractive interaction with the surface atoms (the latent heat of condensation). However, after depositing the equivalent of one monolayer, we found only $\sim 20\%$ of atoms deposited atop islands got incorporated into the islands via push-out. In simulations with 200 ps intervals between deposition events, most atoms were incorporated through thermally activated exchange descent. As the incident atom energy is increased to 10 and 20 eV, characteristic of sputter deposition experiments [4], it is expected that push-out events will become more important, and this was the motivation for the present studies.

Gilmore and Sprague [7,8] have carried out molecular dynamics (MD) simulations of vapor and sputter deposition of Ag atoms on a Ag(111) surface using an embedded atom method (EAM) interaction potential. After depositing the equivalent of several monolayers, they found that an incident atom energy of 0.1 eV resulted in multilayer growth while an incident atom energy of 10 eV resulted in layer-by-layer growth. They inferred that atoms deposited with the higher kinetic energy can disrupt unstable atom configurations to improve film growth. The 10 eV atoms had sufficient energy to displace several film atoms and, although rare, substrate atoms. In subsequent studies where incident atom energies were 10, 20, and 40 eV, Gilmore and Sprague [9] found increased interface mixing occurring by an exchange process between the incident atom and a surface atom. They found that interface mixing was thermally assisted and ballistically activated. In the 10 eV case no interface mixing resulted when the substrate temperature was reduced to 30 K. However, for incident energies of 20 and 40 eV, reducing the substrate temperature did not eliminate the interface mixing, indicating that atoms deposited with these energies were capable of exchanging places with surface atoms without the assistance of thermal fluctuations.

We have conducted a systematic study of atom deposition to identify the various atomic processes that occur. We have carried out a random sampling of 100 impact parameters for sites atop a heptamer and straight type A and B steps (see Fig. 1) to correlate the probability of these exchange events with increased incident atom energy and the region of impact atop an island. In addition to push-out events, we have monitored deposition events resulting in film (or island) atoms being displaced into the substrate or film atoms being displaced atop the growing island, which would result in undesirable effects such as interface mixing and multilayer growth. We have further characterized the exchange processes by finding the minimum energy paths and, thereby, the activation energy barriers using the nudged elastic band method (NEB) [10,11]. We have also studied the effect of small thermal fluctuations, which can have two different kinds of effects. First

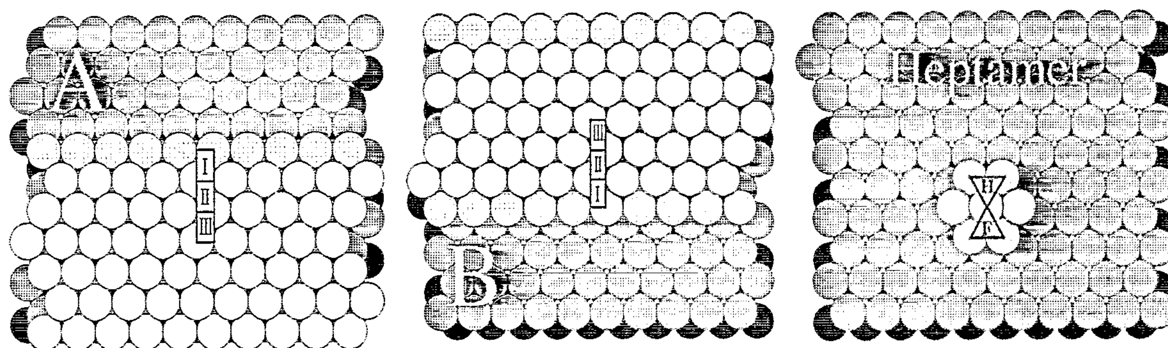

Fig. 1. Schematic of the regions sampled atop A and B type steps and a heptamer. Regions H and F on a heptamer correspond to hcp and fcc stacking positions atop the heptamer. The darker atoms lie lower. The dotted atoms at step and island edges are 0.05–0.1 Å lower than atoms shown with open circles.

of all, the addition of thermal energy may be sufficient to help the system overcome activation energy barriers for some of the exchange processes. The addition of thermal energy could also impede exchange processes involving multiple atoms since the thermal fluctuations could lead to displacement of adjacent atoms which otherwise are aligned to allow efficient momentum transfer for an exchange process.

2. Methodology

We use an EAM-type interaction potential in our MD simulations, as described previously in more detail [4,11]. The short-range interaction of a Pt dimer agrees well with the sputtering potential of Biersack (in the range of 500–1000 eV) [12]. Atoms are deposited with incident energies corresponding to vapor deposition (0.25 eV) and sputter deposition (10 and 20 eV) [6]. The simulations have been carried out at different substrate temperatures, 0 and 275 K, to study the effects of thermal fluctuations. The systems consist of a substrate of between 800 and 1400 atoms, with layers of 100 atoms each, where the bottom two layers are held fixed to prevent translation of the lattice as atoms are deposited. Periodic boundary conditions are used parallel to the surface.

Fig. 2 shows the evolution of the 'hot-spot' at the site of impact, where a deposited atom has incident energy of 20 eV, and the substrate temperature, T_s, is 275 K. (Inset shows the hot-spot evolution for incident energy of 10 eV and $T_s = 275$ K.) The average kinetic energy of the deposited atom and four underlying atoms (in two layers) was calculated over intervals of 0.125 ps. The solid line in Fig. 2 gives the kinetic energy averaged over five runs, each indicated as points. As found in simulations of Pt vapor deposition [4], the excess kinetic energy resulting from a deposition event dissipates very quickly, within 2–3 ps. Such rapid dissipation of the excess energy has been observed previously and analyzed for a simple model system [13]. The simulation runs

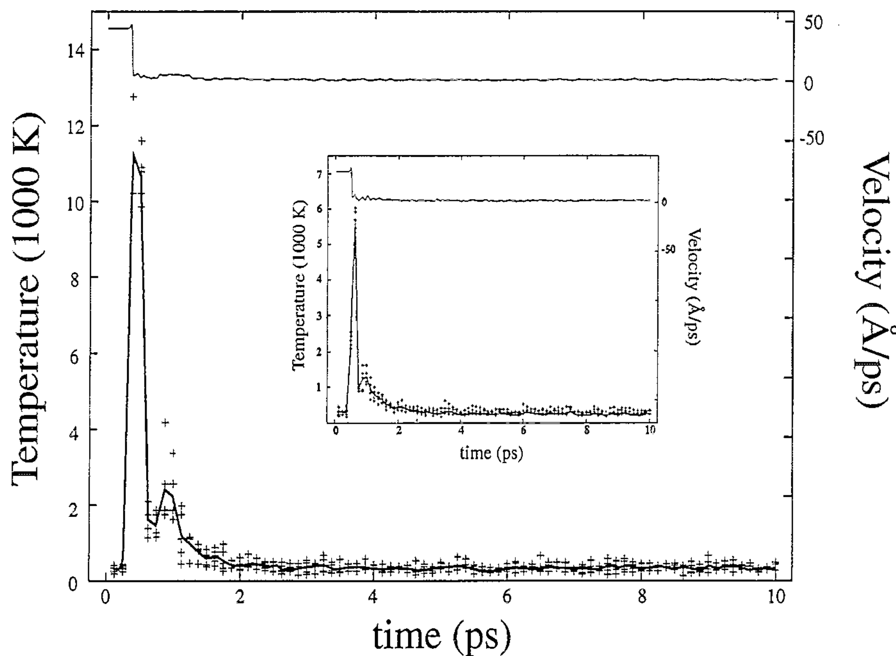

Fig. 2. Kinetic energy (in temperature units) averaged over 5 runs (+) in 12.5 fs intervals within a rectangular region containing the deposited atom and four underlying atoms in two layers below, where the deposited atom has incident energy of 20 eV for $T_s = 275$ K. (Inset given for incident atom energy of 10 eV and $T_s = 275$ K.) The hot-spot cools very quickly, within 2–3 ps for both incident energies. The upper curve shows how the velocity of the impinging atom (scale to the right) is lost immediately after the initial collision with the substrate.

reported here were carried out for a total of 5 ps. Since we are focusing on the effects of increased incident energy for deposited atoms and not effects due to thermally activated diffusion processes, this simulation time is sufficient for full thermal accommodation of the deposited atom, and all the interesting effects pertinent to this study are completed within this time. The upper curve in Fig. 2 (and inset) gives the velocity for the deposited atom and shows how the incoming atom quickly loses its excess kinetic energy upon collision with the surface atoms.

The simulations of the deposition events were done without any temperature control; the system is large enough that the heating due to the deposited atom is small (about 70 K when the incident energy is 20 eV). In our simulation runs, we found identical results for each of the 100 deposition events in one set when the system size was reduced to 800 atoms (bottom two layers held fixed and six fully dynamic layers). Consequently, all simulations were carried out with 800-atom systems for efficiency.

3. Push-out from a heptamer

For a heptamer, there are two types of sites on which a deposited atom may land: the hcp site and the fcc site (designated as H and F in Fig. 1). We sampled 100 randomly chosen impact parameters bounded by the three atoms constituting a stable threefold site for each of the two sites. In the vapor deposition simulation, where the substrate temperature was 0 K and deposited atoms had kinetic energy of 0.25 eV, none of the atoms directed towards region F resulted in a push-out event (see Table 1). Only 17% of the atoms directed towards region H resulted in push-out, where the incoming atom displaced one peripheral heptamer atom to become incorporated at the island edge. These results are consistent with findings from our previous study of the energetics of exchange diffusion processes [11]. We evaluated the activation energy for exchange descent from atop a heptamer and found barriers of 0.48 and 0.29 eV for the fcc and hcp sites respectively. The 0.19 eV barrier height difference can be understood by looking at the position of substrate atoms below the heptamer. While an atom landing on a fcc site displaces a heptamer atom towards an underlying atom, an atom directed on the hcp site pushes a heptamer atom towards a channel, making exchange descent much easier. When we repeated the deposition simulations now for a substrate temperature of 275 K, the probability of push-out from both sites remained practically the same, 0% and 16% for the fcc and hcp sites respectively. These results show that thermal fluctuations do not strongly influence the probability of push-out events from heptamers in vapor deposition.

When we increased the incident energy of the

Table 1
Summary of depositions on the two sites atop a heptamer, hcp and fcc, at $T = 0$ K (275 K)

	[a]	[b]	[c]	[d]	[e]
$E_{inc} = 0.25$ eV					
hcp	17 (16)	0 (0)	0 (0)	0 (0)	83 (84)
fcc	0 (0)	0 (0)	0 (0)	0 (0)	100 (100)
$E_{inc} = 10$ eV					
hcp	54 (38)	20 (28)	0 (0)	0 (0)	26 (34)
fcc	32 (39)	18 (11)	0 (0)	0 (0)	50 (50)
$E_{inc} = 20$ eV					
hcp	26 (19)	54 (61)	0 (0)	2 (6)	18 (14)
fcc	20 (36)	56 (41)	2* (0)	0 (9)	22 (14)

[a]: Singe atom push-out (no atoms atop).
[b]: Multiple atom push-out (no atoms atop).
[c]: Single/multiple* atom push-out leading to island atom atop.
[d]: Multiple atom push-out involving displacement of substrate atom(s).
[e]: Adatom lands atop.

deposited atoms to 10 eV, we found a marked increase in the number of push-out events, including many involving the displacement of several island atoms. Fig. 3 shows the results for deposition on the heptamer at 0 K for the three incident energies 0.25, 10, and 20 eV. The shaded areas correspond to deposition events contributing to smooth film growth, such as push-out of single or multiple island atoms where the deposited atom becomes incorporated in the island. White areas correspond to deposition events where the deposited atom lands atop or where an island atom was displaced to a position atop the island, both of which would contribute to multilayer growth. Fig. 3 shows that an increase in the incident kinetic energy to 10 eV increases the shaded area to include most of the interior of the regions sampled, fcc as well as hcp. As the incident energy is increased the (small) additional barrier for push-out at fcc sites because of the underlying atoms becomes less important, and the two sites give similar results. Mainly those deposited atoms directed to land at the center of a heptamer atom end up atop the heptamer.

In the majority of cases, the 10 eV incident energy is sufficient not only to displace one atom, but to completely disrupt the heptamer, displacing several if not all the island atoms as the deposited atom becomes incorporated. An example of such an event is shown in Fig. 4. We calculated the activation energy barrier for this process using the NEB method [10,11]. Although several atoms are displaced, the NEB is able to determine a minimum energy path and the corresponding activation energy barrier since it requires as input only the initial and final positions of all the atoms in the system. The NEB minimization represents the system as a sequence of images, from the initial state to the final state. All the barrier calculations for this study were carried out using the NEB with 25–30 images. For the exchange process in Fig. 4, the activation energy barrier is 0.69 eV, which is surprisingly low considering three atoms were displaced, including one which was pushed around another island atom. The barrier is so low because the adatom is simultaneously descending and forming three new bonds.

A further increase in the incident atom energy to 20 eV results in a few more push-out events, and a larger fraction of the push-out events involve several atoms (see Table 1). In some cases, the deposited atom had sufficient energy to push a heptamer atom into the substrate, causing the displacement of two

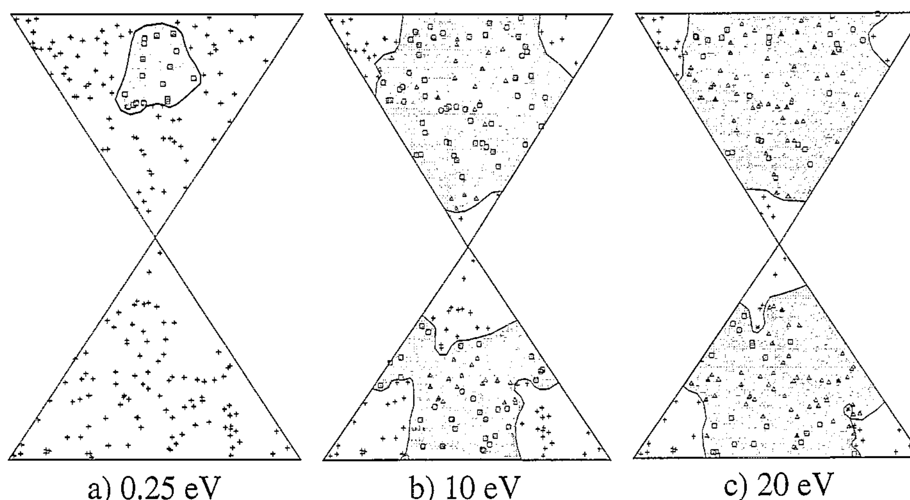

Fig. 3. Schematic of the two regions studied for the heptamer at 0 K and for the three incident energies (a) 0.25 eV, (b) 10 eV, and (c) 20 eV. The shaded areas correspond to deposition events with positive effects, such as push-out of single (□) or multiple (△) island atoms where the deposited atom becomes incorporated in the island. White areas correspond to deposition events where the deposited atom lands atop (+) or where an island atom was displaced to a position atop the island ((×) when a single island atom is displaced and (∗) when multiple island atoms are displaced), both of which would lead to multilayer growth. An increase in the incident kinetic energy from 0.25 to 10 eV increases the area resulting in positive effects to include most of the interior of the regions sampled. Only those deposited atoms directed to land right on a heptamer atom end up atop the heptamer.

substrate atoms, one of which moved out of the substrate to become incorporated at the island edge (see Fig. 5). The barrier for this exchange process was found to be 0.80 eV. For $T_s = 0$ K, one out of a hundred deposition runs resulted in such a displacement of substrate atoms. When the substrate temperature was increased from 0 to 275 K, the probability for this type of push-out increased from 1% to 7.5%, indicating that thermal fluctuations assist the process. To a small extent, increasing the incident energy to 20 eV introduces detrimental effects, such as the potential for interface mixing and displacement of a heptamer atom to atop the island which contributes to multilayer growth. In one event, the deposited atom displaced a heptamer atom to a position atop the island. This type of process contributes to multilayer growth since atoms collecting atop growing islands can combine to form second layer islands before the first layer is complete.

4. Push-out from straight type A and B steps

We also sampled regions of impact at the edge of both type A and B steps (see Fig. 1). The two types of steps have quite different properties [4,11]. At the A type step, an underlying atom blocks an edge atom from moving out perpendicular to the step edge while underlying atoms at a type B step provide a channel whereby atoms can move directly away from the edge. For atoms with incident energy of 0.25 eV and $T_s = 0$ K, there were only six push-out events for the A type step but 22 push-out events for the B-type step (see Tables 2 and 3). These results are consistent with previously determined activation energy barriers for exchange descent from straight steps; the barrier is 0.30 eV at the A step compared with 0.18 eV at the B step [11]. When the substrate temperature was increased to 275 K, only one push-out event resulted for each of the two step types. This can be understood by noting that at $T_s = 0$ K,

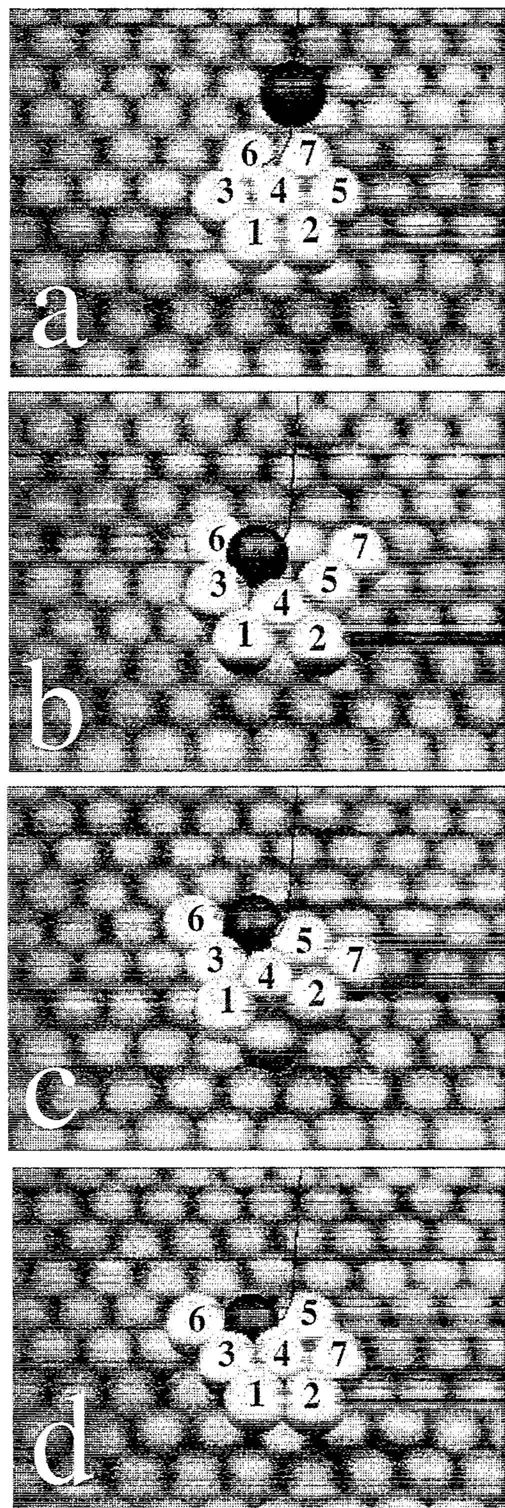

Fig. 4. Sequence of events showing the incorporation of a 10 eV incident atom (black) into a heptamer (white) by displacement of several heptamer atoms sitting on a (111) substrate (grey). The calculated activation energy for this process is 0.69 eV. This type of process would be conducive to layer-by-layer growth.

Table 2
Summary of depositions on regions I–III atop a type A step (see Fig. 1), at $T = 0$ K (275 K)

	[a]	[b]	[c]	[d]	[e]
$E_{inc} = 0.25$ eV					
I	6 (1)	0 (0)	0 (0)	0 (0)	94 (99)
II	0 (0)	0 (0)	0 (0)	0 (0)	100 (100)
III	0 (0)	0 (0)	0 (0)	0 (0)	100 (100)
$E_{inc} = 10$ eV					
I	45 (50)	12 (14)	(0)	0 (0)	43 (36)
II	0 (0)	32 (23)	0 (0)	0 (0)	68 (77)
III	0 (0)	0 (12)	0 (0)	0 (0)	100 (88)
$E_{inc} = 20$ eV					
I	58 (66)	16 (20)	1 (0)	0 (0)	25 (14)
II	0 (1)	58 (57)	11 (0)	0 (0)	31 (42)
III	0 (0)	17 (3)	13 (6)	0 (0)	70 (91)

Categories [a]–[e] are as detailed in Table 1.

underlying atoms at a B type edge are optimally positioned to create a channel by which edge atoms can move directly away from the step in an exchange process. The addition of thermal fluctuations disrupts this alignment and impedes the displacement of the type B edge atoms. For the A type step, an underlying atom blocks an edge atom from moving directly outwards and this is probably not affected appreciably by the small thermal fluctuations. The slight drop in probability of push-out from 6% to 1% with increased temperature is probably not statistically significant.

Surprisingly, for the B type step, the one push-out event was a single atom displacement after deposition on region II. After landing atop the island, the deposited atom diffused to the edge where it underwent exchange with a step atom. For this deposition event, the deposited atom experienced a reversal in its perpendicular momentum but still effected an exchange descent process. In our previous vapor deposition simulation [4], we distinguished between push-out effects and thermal exchange descent processes by assigning the push-out label only to those events where the deposited atom experienced no reversal in its perpendicular momentum before incorporation into a growing island. In the present study, we have broadened the push-out term to include all exchange events directly related to a deposition event

Table 3
Summary of depositions on regions I–III atop a type B step (see Fig. 1), at $T = 0$ K (275 K)

	[a]	[b]	[c]	[d]	[e]
$E_{inc} = 0.25$ eV					
I	22 (0)	0 (0)	0 (0)	0 (0)	78 (100)
II	0 (1)	0 (0)	0 (0)	0 (0)	100 (99)
III	0 (0)	0 (0)	0 (0)	0 (0)	100 (100)
$E_{inc} = 10$ eV					
I	55 (55)	2 (5)	0 (4)	0 (0)	43 (36)
II	1 (0)	19 (32)	0 (0)	0 (0)	80 (68)
III	0 (0)	0 (0)	0 (0)	0 (0)	100 (100)
$E_{inc} = 20$ eV					
I	65 (68)	3 (7)	5 (2)	2 (0)	25 (23)
II	4 (4)	46 (34)	11 (2)	2 (1)	37 (59)
III	0 (0)	15 (1)	18 (6)	0 (0)	67 (93)

Categories [a]–[e] are as detailed in Table 1.

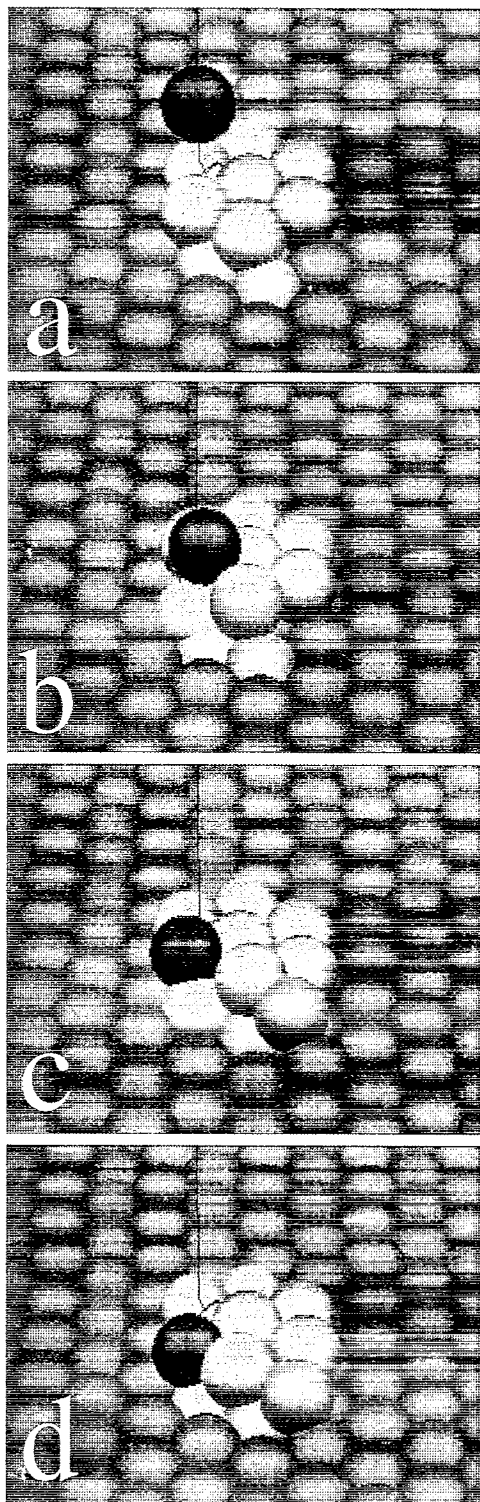

Fig. 5. Sequence of events showing the incorporation of a 20 eV incident atom (black) into a heptamer (white) by displacement of a heptamer atom into the substrate, causing one substrate atom to move out and become incorporated at the island edge. The calculated activation energy for this process is 0.80 eV. This type of process would lead to interface mixing.

and the resulting hot-spot such that the exchange process is completed within 5 ps. We have monitored the trajectories of the atoms deposited onto the type B step in region I. The number of trajectories where the incoming atom pushes into the island without reversal of momentum increases from 56% to 68% as the incident energy is increased from 10 to 20 eV. This is not surprising as atoms with higher incident energy can displace island atoms more easily in the first collision.

Fig. 6 shows the three regions studied for the type A step at $T_s = 0$ K and for the three incident energies 0.25, 10, and 20 eV. Again, shaded regions correspond to deposition events enhancing layer-by-layer growth, where the deposited atom was incorporated in the island, and white regions correspond to deposition events contributing to multilayer growth, where the adatom landed atop the island or displaced an island atom to a position atop the island. Increasing the adatom kinetic energy to 10 eV greatly increases the area of single atom push-out in region I and introduces multiple atom push-out in regions I and II (see also Table 2). Fig. 7 shows a multiple atom push-out where the deposited atom displaces two island atoms near the edge of an A type step, with the final configuration (Fig. 7b) showing an island atom protruding at the step edge. We calculated an activation energy barrier of 0.60 eV for this process. When the incident energy is increased to 20 eV (Fig. 6c), a larger area for single and multiple atom push-outs in regions I and II is found, and push-out even becomes possible in region III. This indicates that near layer-by-layer growth in sputter deposition can be achieved when islands are small enough, with a width of seven or fewer atoms.

The 20 eV incident energy, however, also results in some less desirable effects. Fig. 8 shows a deposition event where an atom deposited at a type B step displaces an edge atom to a position atop the island. The activation barrier is 0.73 eV. These types of

events would be detrimental in thin film growth as they would result in atoms collecting atop an island, promoting multilayer growth. Still, Fig. 6 indicates that deposition events resulting in push-out to positions atop the island in regions II and III are concentrated in areas that resulted in deposited atoms landing atop for $E_{\mathrm{inc}} \leqslant 10$ eV, so this negative effect does not detract from the larger regions of impact parameters that resulted in the incorporation of the incident atom for $E_{\mathrm{inc}} = 20$ eV. The results for the type B step were essentially the same, with a greater number of positive push-out events with increasing incident atom energy.

As discussed earlier in the vapor deposition simulations, increasing the substrate temperature to 275 K greatly reduces the number of push-out events in region I for both type A and B steps (see Tables 2 and 3). Incident energies of 10 and 20 eV are sufficiently high that an increase in the substrate temperature does not have a significant effect (see Tables 2 and 3).

In two of the 20 eV deposition events at straight steps, we observe the formation of a bulk vacancy, where the deposition event caused a substrate atom to be displaced and become incorporated in the island. The activation barrier for this process is very

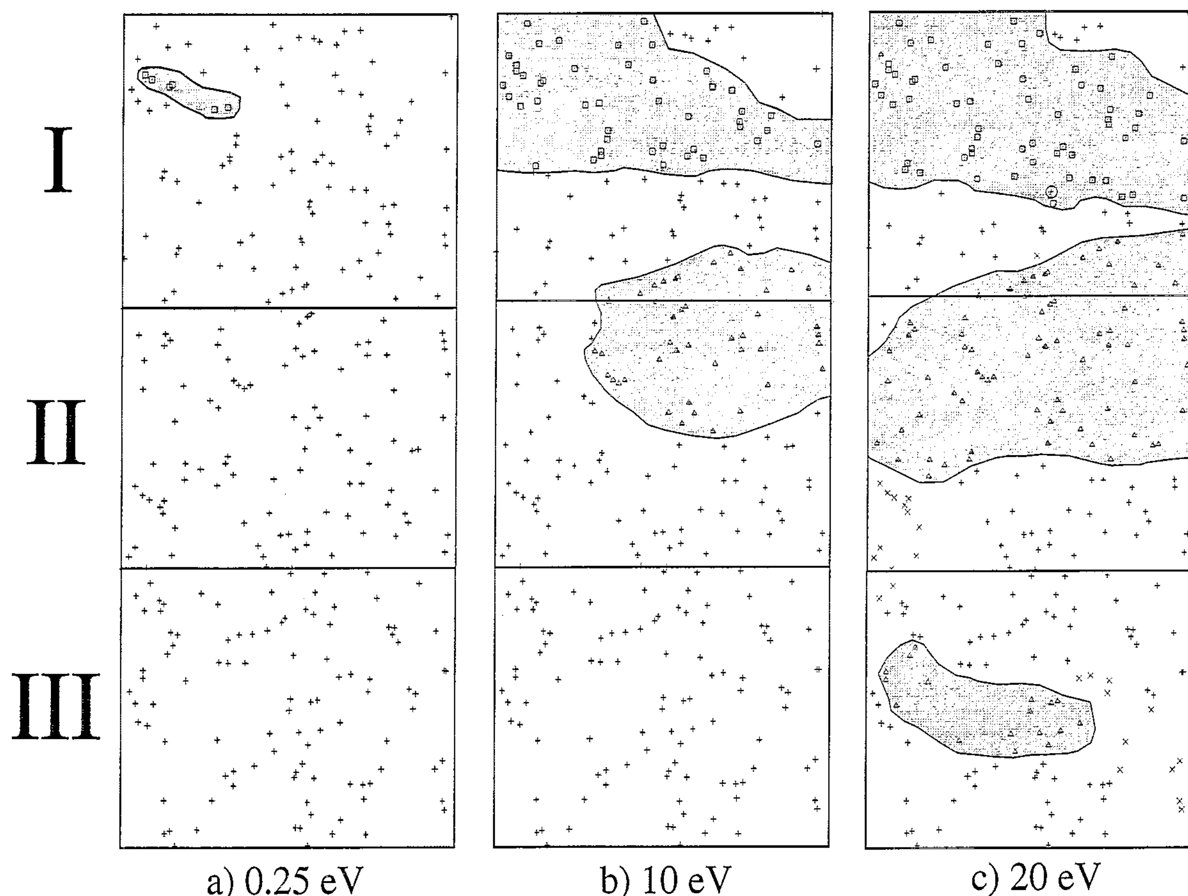

Fig. 6. Same as Fig. 3 but for the three regions studied at the type A step at 0 K (see Fig. 1), also for the three incident energies (a) 0.25 eV, (b) 10 eV, and (c) 20 eV. Increasing the incident atom energy from 0.25 to 10 eV increases the number of single atom push-out events in region I and introduces multiple atom push-out events in regions I and II. An increase in the incident energy to 20 eV further increases the areas leading to layer-by-layer growth into region III. The 20 eV kinetic energy also enables the deposited atom to displace single (×) and multiple (∗) island atoms such that one island atom comes to rest atop the island.

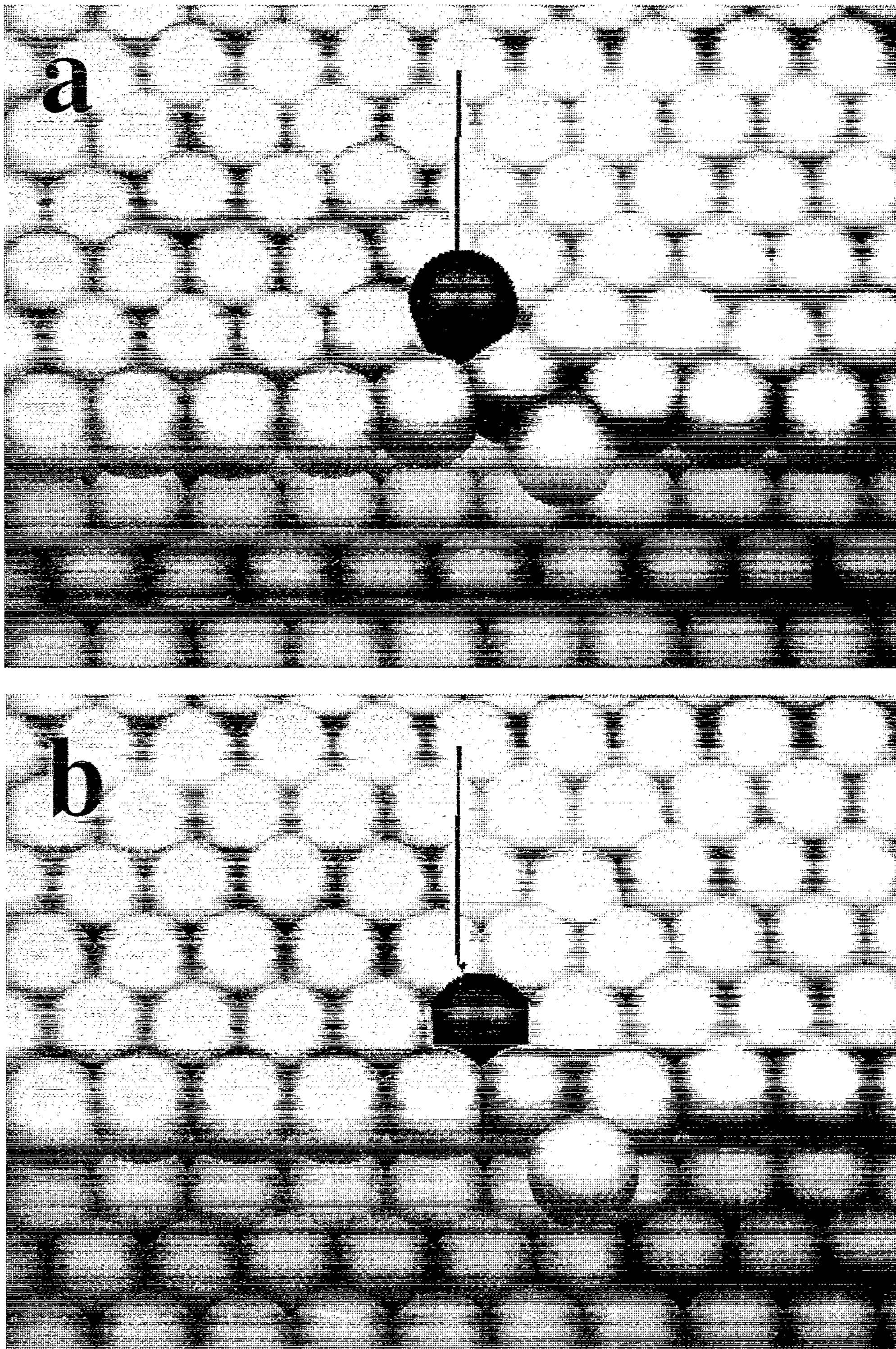

Fig. 7. Sequence of events showing the incorporation of a 10 eV incident atom (black) at a type A step (island atoms in white) by displacement of two island atoms. The calculated activation energy for this process is 0.60 eV. This type of process would promote layer-by-layer growth.

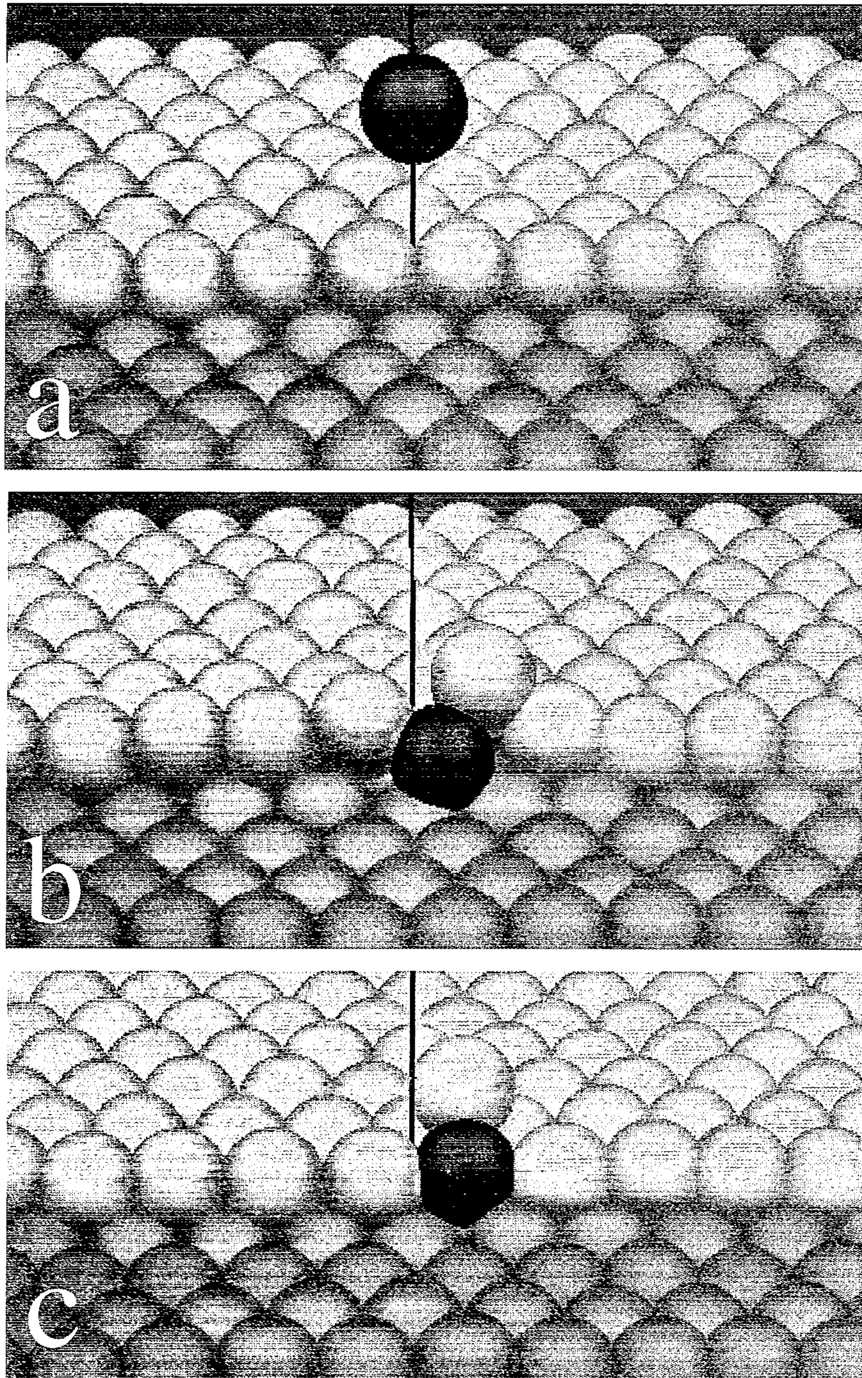

Fig. 8. Sequence of events showing the incorporation of a 20 eV incident atom (black) at a type B step (island atoms in white) by displacement of one island atom to a position atop the island. The calculated activation energy for this process is 0.73 eV. This type of process would promote multilayer growth since an atom comes to rest atop the island.

high (2.36 eV) mostly because the final state is 1.97 eV higher in energy than the initial state. The high activation energy barrier makes vacancy formation a very rare event and its instability makes it susceptible to annealing in subsequent depositions, but such events are worth noting since the presence of vacancies would greatly reduce the quality of a thin film.

5. Conclusions

By raising the incident kinetic energy of deposited Pt atoms from 0.25 eV, consistent with vapor deposition, to 10 and 20 eV, consistent with sputter deposition, an increasing area of impact atop a heptamer and straight type A and B steps on Pt(111) results in push-out events where incident atoms get incorporated into the growing layer. These results indicate that near layer-by-layer growth can be expected when islands on the surface are small enough (seven or fewer atoms in diameter) and incident atom energies are on the order of 10–20 eV. The probability of push-out events at B type edges can also be enhanced by lowering the substrate temperature in vapor deposition since thermal fluctuations tend to disrupt a channel formed by the underlying atoms allowing an edge atom to be displaced directly away from the edge. This thermal effect is not significant when the incident energy is 10 eV or higher.

To some extent, the 20 eV incident energy also results in exchange with surface atoms and even displacement of subsurface atoms which would lead to interface mixing in heteroepitaxial growth. Lowering the incident energy to 10 eV eliminated these events in the present studies.

We have evaluated the activation energy barriers for several representative displacement mechanisms. The barriers were found to be surprisingly low (0.6–0.8 eV) even for exchange processes involving multiple atoms, with the highest barrier corresponding to a process where in addition to an island atom two substrate atoms were displaced.

Acknowledgements

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