Height-dependent barriers and nucleation in quantum size effect growth

Z. Kuntová,[1] M. C. Tringides,[2] and Z. Chvoj[1]

[1]*Institute of Physics, Academy of Sciences of the Czech Republic, v. v. i., Na Slovance 2, 182 21, Prague 8, Czech Republic*
[2]*Ames Laboratory US DOE, Iowa State University, Ames, Iowa 50011, USA*

An analytic model is developed to model the growth of the uniform height islands. The formation of bilayer rings on top of stable islands and the absence of nucleation on top of the third layer are particularly intriguing. The analysis is motivated from recent Monte Carlo simulations that have reproduced the bilayer morphology. The analytic model allows better transparency to the role of the different barriers. In particular the diffusion anisotropy (i.e., atoms diffuse faster azimuthally within the ring than toward the island center) explains why nucleation is only observed close to the island edge and not in the middle. Lower barriers to diffuse back to the wetting layer from unstable heights than the barriers from stable heights explain the absence of nucleation on the third layer. The model can have general use in other systems where quantum size effects play a role and the barriers become height dependent.

I. INTRODUCTION

One of the unusual systems that has generated a lot of interest recently is Pb/Si(111). Numerous experiments by different techniques [spot-profile analysis of low-energy electron diffraction (SPA-LEED),[1] scanning tunneling microscopy/scanning tunneling spectroscopy (STM/STS),[2,3] x-ray,[4,5] angle-resolved photoemission spectroscopy (ARPES),[6,7] *in situ* conductivity,[8] etc.] and corresponding theoretical calculations[9,10] have focused on the role of quantum size effects (QSE) on island height stability. Unusually sharp height distributions have been observed where a single height is selected. This stability has been attributed to the special relation between the Fermi wavelength λ_F and inter-layer spacing d for Pb(111) $d \approx (3/4) \lambda_F$. However the nucleation and the kinetics of island height formation raise important questions about novel diffusion mechanisms at low temperatures and nonclassical effects on mass transport.[11–13] Coarsening experiments[5] have shown that faster coarsening times are observed during growth at high than low flux rates because a large fraction of unstable heights form initially that easily decay and transfer material to the stable heights. Other studies have shown that the kinetic barriers are height dependent,[11,13] with both the terrace diffusion barrier and the step edge barrier dependent on whether the island height is stable or unstable one. An additional important process is related to the high density of the wetting layer [in excess of the density of metallic Pb(111)] that moves collectively from the region outside to the island top.[12]

Despite this large number of unusual experimental findings theoretical modeling of the nucleation and growth of the uniform height islands still leaves a lot of questions unanswered. One question is how is it possible to nucleate bilayer rings which have smooth inside boundaries when the initial island height is a stable one (and single layer rings when the initial height is an unstable one). Although these bilayer rings are clearly favored energetically because they avoid the unstable layers it is still not clear what the key controlling barriers are and how they depend on QSE. Identifying these kinetic processes is essential since the formation of the uni-

form height islands occurs over a narrow window of temperature and coverage growth parameters. It can guide the search of finding the optimal window in other systems as well.

Earlier work with Monte Carlo simulations[14,15] has identified some key barriers that are responsible for this unusual growth, in particular, the anisotropy of diffusion with the incoming Pb atoms diffusing much slower out of one lattice constant ring than diffusing azimuthally within the ring. This enhances nucleation preferably at the island edge. It is also essential to have a very low diffusion barrier consistent with first-principles calculations[11] that suppress nucleation in the middle of the terrace. Another important barrier difference was the one controlling transfer from the island top back to the wetting layer with the barrier for unstable height lower than the one for stable heights. The simulation reproduced the observed bilayer rings as seen in the experiment but the results were not very transparent. The goal of the current paper is to carry out analytic work to model the nucleation process on the different heights and to identify why the nucleation of the third layer is suppressed and only bilayer rings are seen.

II. MODEL

Although the current model has certain simplified assumptions, it captures the main features of the very unusual uniform height island growth observed in experiment.[1] These unusual features are seen in Fig. 1 where an ~120 nm Pb island was grown on the Si(111)-7×7 displaying a bilayer smooth ring at 240 K. The height is measured from the wetting layer (not the Si substrate). The grown islands have preferred odd heights of five, seven, and nine layers and the islands with heights of six, eight, and ten layers are rarely observed. The odd heights are the stable layers and the even heights are the unstable layers. Monolayer rings are observed on unstable heights. Unstable islands are grown at higher temperature than ~240 K and become laterally big (more than 200 nm). They grow to the next stable height instead of decay. For growth on stable heights no nucleation is ob-

FIG. 1. (Color online) 281×205 nm^2 STM image of the Pb islands grown on Si(111)-(7×7) surface. The numbers indicate the height of the different areas and a smooth bilayer ring has formed.

served on the eighth layer as seen in Fig. 1 before both lower layers are filled.

Both heights of five and seven layers are stable heights on the 7×7 but the seventh layer is a superstable height, i.e., after a sufficiently long time depending on the initial island size practically all islands convert to seven-layer heights. The Pb material needed to convert unstable into stable islands is coming from the wetting layer to the top layer of island. As discussed elsewhere this corresponds to annealing experiments or deposition experiments at higher temperatures and with minimal amount $\Delta\theta < 1$ ML deposited.

The current model describes the transfer of atoms from the wetting layer. No deposition of atoms directly on top of the island is assumed. This process was modeled by the kinetics of atoms on stable or unstable layers within a lattice gas model. Each layer is constructed with lattice sites (occupied by atoms or unoccupied) which form a hexagon with the length of its side R (in units of interatomic distance). [In the real crystal the radius of the hexagon in the upper layer should be smaller by one lattice constant than the radius of the lower layer because of the ABC stacking of the fcc(111) crystal. But in the analytic model we neglect this and we assume that R is the same for all layers.] We distinguish stable layers (5, 7, 9) and unstable layers (6, 8) with different energy barriers according to the results of the recent Monte Carlo simulations[14,15] that provided an optimal set of barriers that reproduce the observations. From the simulation the following conclusions were reached. The diffusion of adatoms is highly anisotropic on each layer. There exists one lattice constant wide ring $\mathfrak{R}$ on the edge of a layer with low azimuthal diffusion barrier within the ring or anywhere outside the ring, E_t. The diffusion barrier for jumps of single atoms from the ring toward the center is much higher, $E_b > E_t$. This assumption was based on the empirical observation in controlled experiments that the ring spreads out azimuthally much faster than radially.

It is not clear what the physical origin of the diffusion anisotropy is and how it is related to QSE. However recent work has shown that QSE modify the terrace diffusion barrier for stable vs unstable islands[11] so it is possible that the barrier is also modified at the island edge vs center. In addition in Ref. 11 it was shown theoretically that there must be preference for atoms on unstable heights to adsorb preferably within a ring close to the island perimeter. This evidence supports the assumption that there must be diffusion anisotropy for azimuthal vs radial diffusion although full justifi-

cation of this assumption requires first-principles calculations of the diffusion barriers at the perimeter vs island center. The separability of the confined electron wave function (into normal and lateral components) is better met at the island center and therefore the QSE standing wave condition is better justified. Close to the island perimeter the electron wave function can leak out both in the normal and lateral direction and smear out the standing wave condition. This implies that the energy cost to nucleate at the edge is lower than at the island center. There must be a relation between the diffusion anisotropy (azimuthally vs radially within the ring) and the differences in the confined electron wave functions close and away from the island perimeter.

Because of the difference in the diffusion barrier and because the source of atoms is the wetting layer the density of atoms inside the ring is ρ_b and outside the ring ρ_r are very different, $\rho_r \ll \rho_b$. The densities ρ_b ρ_r are defined as the number of atoms divided by the number of adsorption sites. Within this definition these densities are dimensionless and mean the relative occupation with respect to saturation. The value is within the interval (0,1). We define the number of atoms i_b in critical cluster; i.e., once a cluster reaches this size it never falls below it by atom detachment. The formation of the critical cluster is a stochastic process and the probability of a cluster to appear is time dependent. We investigate the nucleation probability within the ring (i.e., when the critical cluster has at least one atom within the ring). As will be shown later the most probable nucleation is when all the atoms of the critical cluster are within the ring. Nucleation toward the island center is suppressed (despite the larger available area) because the density ρ_r is much less.

III. KINETIC PROCESSES AND FLUX RATES OF ADATOMS IN ISLAND HEIGHT GROWTH

The kinetics of atoms on island surfaces is determined by the probabilities of jumps of atoms from the wetting layer to the island top, the opposite jumps from the top back to the wetting layer, jumps to the lowers layer and jumps between different adsorption sites on the top of the island. These probabilities follow Arrhenius form, which implies that they are determined by the prefactors and energy barriers. We show schematically in Fig. 2 the different microscopic processes and parameters of the model.

All the prefactors v were taken as one for all the process which means the time unit τ_0 is the inverse of the prefactor v^{-1}. Within this choice, the wetting layer atoms can jump on the top layer with the probability P_{WL}:

$$P_{\mathrm{WL}} = \exp\left(-\frac{E_{\mathrm{WL} \to t}}{kT}\right), \qquad (1)$$

where $E_{\mathrm{WL} \to t}$ is the energy barrier, T is temperature, and k is the Boltzmann constant.

For each barrier we include in addition to the Boltzmann factor the number of nearest-neighbor sites available for the atom to hop into: the number of sites on the terrace is six to the next-nearest positions, the number of sites to jump from the wetting layer is two. These normalization factors give the
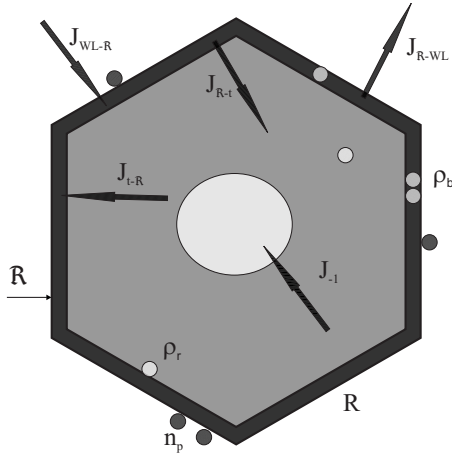
FIG. 2. The geometrical model of the island top with all the contributing fluxes of atoms and the relevant model parameters.

probability to jump on the terrace or azimuthally within the ring:

$$P_t = \frac{1}{6}\exp\left(-\frac{E_t}{kT}\right). \tag{2}$$

From the energy barriers and these normalization factors we can write expressions for the different fluxes in and out of the ring.

First the flux of atoms from wetting layer to the top layer. We introduce dimensionless parameter n_p, which measures the effective two-dimensional (2D) deposition rate (as a result of the arrival rate of the wetting layer to the island perimeter). Atoms are incident on the unit length of the island perimeter from the wetting layer, and jump on the top layer with probability P_{WL}. The flux of atoms $J_{WL-\Re}$ from the wetting layer to the island top is

$$J_{WL-\Re} = 6Rn_p \exp\left(-\frac{E_{WL\to t}}{kT}\right)(1-\rho_b), \tag{3}$$

where ρ_b is density of atoms within the ring $\Re$. The blocking factor $(1-\rho_b)$ expresses the decrease of the flux $J_{WL-\Re}$ to the ring with increasing occupation of the ring sites.

The flux $J_{\Re-WL}$ of atoms from the ring $\Re$ to the wetting layer is given by the relation

$$J_{\Re-WL} = 2R\rho_b \exp\left(-\frac{E_{t\to WL}}{kT}\right), \tag{4}$$

where $E_{t\to WL}$ is energy barrier for the jump of atoms from ring to the wetting layer. The energy barrier is assumed to be different for atoms on the stable layer and on the unstable layer. There are $6R\rho_b$ atoms to hop from the top back to the wetting layer. An atom in the ring can only jump along two directions since the ring is of one lattice constant width. Since the fluxes are realized by the jumps of atoms in two directions (from the six possible directions two are inside the ring, two are toward the terrace away from the ring, and two toward back the wetting layer) it is only 1/3 of all possible jumps. Thus prefactor is 6/3=2. The difference between stable and unstable layers is reflected in the barrier $E_{t\to WL}$.

Since we balance total fluxes on and out from the island, we must consider atom input to the wetting layer from atoms in the sixth, seventh, and eighth layers. Atoms on the sixth layer can jump back to the wetting layer with barrier $E_{t\to WL}(6)$. However the only atoms on the sixth layer which can jump to the wetting layer are the ones that do not have seventh layer atoms directly above them. $6R\rho_b(6)$ is the number of atoms within the ring in the sixth layer and $6R\rho_b(7)$ is the number of atoms within the ring in the seventh layer and also directly above sixth layer atoms. This implies that only $6R[\rho_b(6)-\rho_b(7)]$ atoms can jump from the sixth layer to the wetting layer. A similar argument works for atoms on the seventh layer because we allow partial occupation of the eighth layer. (For the eighth layer no ninth layer atoms are to be present assumed as in the experiment.) If we multiply this number of atoms jumping from the ith layer by probability of jump per unit time $\exp[-E_{t\to WL}(i)/kT]$ and multiply it by factor 1/3 (only one third of possible jumps on the hexagonal lattice are in the direction of the wetting layer), we obtain the particular flux from ith layer. The sum of these fluxes is for $i=6$ or 7 or 8:

$$J_{WL}^{tot} = 2R\{[\rho_b(6) - \rho_b(7)]\exp[-E_{t\to WL}(6)/kT]$$
$$+ \{\rho_b(7) - \rho_b(8)\exp[-E_{t\to WL}(7)/kT]$$
$$+ \rho_b(8)\exp[-E_{t\to WL}(8)]/kT\}\}.$$

If we reorganize terms with the same exponential function and put energy barriers $E_{t\to WL}(8)=E_{t\to WL}(6)$ as in the table, we obtain the following equation:

$$J_{WL}^{tot} = 2R\left\{\rho_b(6)\exp\left[-\frac{E_{t\to WL}(6)}{kT}\right] + [\rho_b(7) - \rho_b(8)]\right.$$
$$\left. \times\left\{\exp\left[-\frac{E_{t\to WL}(7)}{kT}\right] - \exp\left[-\frac{E_{t\to WL}(6)}{kT}\right]\right\}\right\}. \tag{5}$$

We explicitly write the dependence of $E_{t\to WL}(h)$ on island height $h=6$ or 7.

There are the following interesting special cases: $\rho_b(7)=\rho_b(8)=0$—growth of the sixth layer $\rho_b(6)<1$, $\rho_b(8)=0$ $\rho_b(6)=\rho_b(7)=1$ growth of bilayer, no nucleation on eighth layer (as seen in Fig. 1). The flux $J_{\Re-t}$ from the ring in the other direction toward the island center is given by

$$J_{\Re-t} = 2R\rho_b \exp\left(-\frac{E_b}{kT}\right)(1-\rho_r), \tag{6}$$

where ρ_r is the density of atoms on the terrace and it can differ for stable and unstable layers. The flux $J_{t-\Re}$ of atoms from the outside of the ring on the island to the ring $\Re$ is given by

$$J_{t-\Re} = 2(R-1)\rho_r \exp\left(-\frac{E_t}{kT}\right)(1-\rho_b). \tag{7}$$

The flux J_{-1} of atoms from a top layer to a lower layer (e.g., from eighth layer to seventh or sixth layer) is given by

TABLE I. Table showing the barriers identified in Ref. 15 that were used in the current model to estimate the ring and hole filling time on the different layers.

Barrier	Value	Process
E_b	0.17eV	Energy barrier for jump from the ring $\Re$ toward center
E_t	0.05eV	Energy barrier for diffusion on terrace
$E_{t \to \mathrm{WL}}$	0.32eV	Energy barrier for hopping from the top of stable island to WL
$E_{t \to \mathrm{WL}}$	0.21eV	Energy barrier for hopping from the top of unstable island to WL
$E_{7 \to 6}$	0.2 eV	Energy barrier for falling of atom from seventh to sixth layer
$E_{\mathrm{WL} \to t}$	0.25eV	Energy barrier for "climbing" up from WL to the top of island
i_b	8	Size of critical nucleus
n_p	5	Parameter describing the deposition flux intensity
T	180 K	Temperature
R	30	Radius of the island in the number of atoms

$$J_{-1} = 2r\rho_r \exp\left(-\frac{E_t + E_{i \to j}}{kT}\right),\qquad (8)$$

if the lower layer is not filled and $E_{i \to j}$ is a measure of any descending barrier that might be present at the edge of the inside of the ring.

In summary we use six barriers to describe the transfer of atoms between the different layers and the different regions of the island which were identified in the previous Monte Carlo simulations. The values of these barriers are shown in Table I. The model of surface potential is shown in the schematic diagram of Fig. 3.

IV. DETERMINATION OF MODEL PARAMETERS

The main question is to determine the probability of the formation of a new stable cluster on the ring $\Re$ as a function of layer. First the time of the filling of each layer can be estimated from the difference of fluxes $J_{\mathrm{WL}\text{-}\Re} - J_{\mathrm{WL}}^{\mathrm{tot}}$. We denote the number of atoms to fill 1 ML (or 2 ML for the case

of bilayer growth) layer N_M. The time t_c needed for the filling of a layer is

$$N_M = \int_0^{t_c} [J_{\mathrm{WL}\text{-}\Re} - J_{\mathrm{WL}}^{\mathrm{tot}}(t)]dt = J_{\mathrm{WL}\text{-}\Re}t_c - \int_0^{t_c} J_{\mathrm{WL}}^{\mathrm{tot}}(t)dt.$$

$$(9)$$

In this equation $J_{\mathrm{WL}\text{-}\Re}$ is assumed constant and independent of layer. The flux of atoms from the wetting layer has a constant, time-independent part, multiplied by the blocking factor $(1 - \rho_b)$. This factor implies a decrease in the flux from the wetting layer to the top with increasing ρ_b. Our analysis applies up to the onset of nucleation. It corresponds to the critical density $\rho_b \approx 0.1$. Up to this density the blocking factor can be approximated by 1, so we can assume that the flux of atoms from the wetting layer is time independent. $J_{\mathrm{WL}}^{\mathrm{tot}}$ is the flux out of the top of the island and depends on the critical density $\rho_b(t)$ (which is time dependent). Since we study the ability of the system to nucleate on the ring $\Re$, we find the lowest bound for this so the completion time can be approximated as

$$t_c \approx \frac{N_M}{J_{\mathrm{WL}\text{-}\Re}}.\qquad (10)$$

The assumption behind the estimation is that two competing times are the completion of a layer t_c given by Eq. (10) and the nucleation time t_n. The nucleation time depends on several parameters (the critical size cluster i_b, the barriers and densities of atoms on the ring and terrace). It can be estimated from the probability of nucleation of a critical cluster of size i_b on the ring $\Re$ when the density in the ring has some value ρ_b, at some time t. Since these times to nucleate have a distribution we estimate the mean nucleation time and identify this time with t_n and we compare it with t_c. If $t_n \gg t_c$, nucleation does not realize otherwise nucleation happens prior to the layer completion.

The controlling factors are the difference between the fluxes of atoms to the ring $\Re$ from the wetting layer minus the flux of atoms from the ring $\Re$ back to the wetting layer or toward the inside of the layer. Because of the different barriers used to describe the different layers the nucleation time t_n depends on the layer. As will be shown later the most probable location of the second layer critical cluster is within the ring than close to the center [because the density within
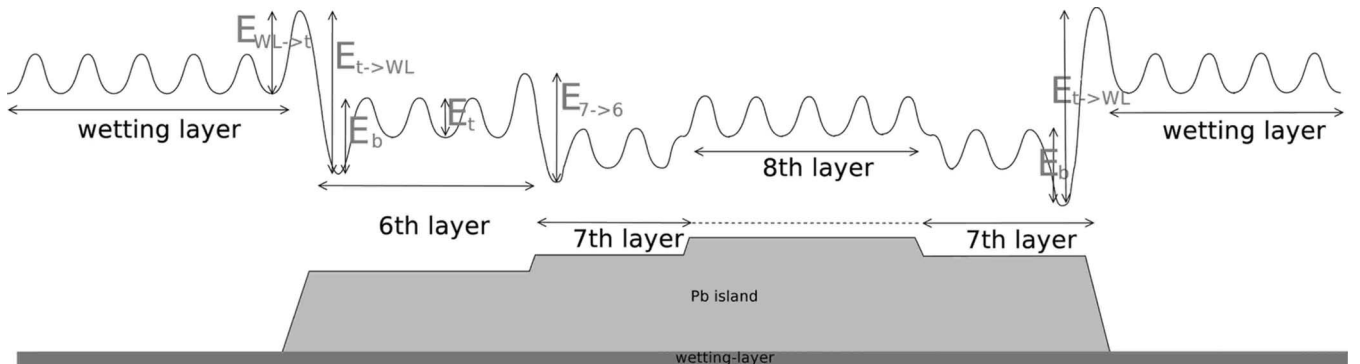

FIG. 3. Schematic diagram illustrating energy barriers used in analysis of nucleation on different layers.

the outside area is low especially for the time that critical nucleation density within the ring $\rho_b^c(i_b)$ is attained].

This model includes several parameters, shown in Table I, which were determined from the experiment. For example $T=180$ K, $R=30$ lattice constants and $n_p=5$ are based from the parameters in an earlier experiment.[16] The factor $n_p=5$ is determined by the 2D flux rate since the diffusion on the wetting layer is extremely fast and the deposited atoms very quickly move to the growing islands. The critical size cluster describing the nucleation process on the ring $\Re$ $i_b=8$ since in Ref. 14 it was found to be larger than 4. The densities $\rho_{r(b)}$ and fluxes J_i are results of the calculations. From these barriers we estimate the rates to jump from the wetting layer $P_{WL}=1\times10^{-7}[\tau_0]^{-1}$, to diffuse on the terrace $P_t=6.6\times10^{-3}[\tau_0]^{-1}$. The number of atoms needed to complete a single layer on top of the islands is $[N_M=3(R+1)R]$ $N_M=2790$ and to complete the bilayer is $N_M=5580$. The rates were calculated for one specific temperature $T=180$ K as an illustration of the competing effects. The temperature is the one used in the experiment of Ref. 16. The rates differ by several orders of magnitude as seen by the competing barriers in Table I (terrace diffusion anisotropy 0.05 eV on the terrace vs 0.17 eV out of the ring, difference for hopping back to the wetting layer 0.21 eV from stable vs 0.32 eV from unstable height). However calculations of the corresponding quantities within the temperature range 140 K $<T<220$ K support the current calculations and predictions about the probability of nucleation at the different layers because of this large variation in barriers. This is the experimental range where the seven-layer uniform height islands are observed.

Using these barriers for individual atom jumps, we can determine the fluxes in and out of the ring which determine whether nucleation is possible in a given layer or not. Depending on the relative rate of the processes either the critical density is attained and nucleation is observed or the density stays below the critical value and nucleation is not observed. First the fluxes to the ring which increase the density ρ_b, the flux of atoms from wetting layer to the top layer, are $J_{WL\text{-}\Re}=9\times10^{-5}[\tau_0]^{-1}$. The flux of atoms from the terrace on top of the island to the ring $\Re$ is $J_{t\text{-}\Re}=2.3\times\rho_r(1-\rho_b)[\tau_0]^{-1}$. These flux rates are competing with the process that removes atoms from the ring $J_{\Re\text{-}WL}$ to the wetting layer. For stable heights $J_{\Re\text{-}WL}=6\rho_b\times10^{-8}[\tau_0]^{-1}$ and for unstable heights it is $J_{\Re\text{-}WL}=8\rho_b\times10^{-5}[\tau_0]^{-1}$. The flux of atoms from the inside of the ring $\Re$ toward the center of the island at the top layer is $J_{\Re\text{-}t}=1\times10^{-3}\rho_b(1-\rho_r)[\tau_0]^{-1}$. The flux of atoms from the seventh to the sixth layer is $J_{-1}=2\times10^{-7}r\rho_r[\tau_0]^{-1}$, and from the eighth to the seventh layer, it is $J_{-1}=8\times10^{-2}r\rho_r[\tau_0]^{-1}$ where r is radius of the hole in the sixth and seventh layer. This radius r is a function of time as the hole radius decreases to zero. The flux of atoms which fall into this hole is proportional of the density of atoms on the higher layer at the edge of hole and on the perimeter of the hole.

V. PROBABILITY OF THE NUCLEATION IN THE RING $\Re$ AS A FUNCTION OF TIME

If the first critical nucleus appears in the system, it implies that a cluster with i_b atoms has formed. Since the diffusion on the terrace is very fast, we neglect the radius dependence of ρ_r: we assume that very quickly the density becomes uniform everywhere in the hole. The surface area of the ring is $6R$ and the area of the rest of the top layer is $S\leq3(R-1)R$.

Hole is the empty area on a particular layer without atoms. It is seen directly in the experiment of Fig. 1. Since the ring is assumed to be one lattice constant the "hole" must have a radius less than $R-1$. It is important to have a hole in the ith layer for atoms from the $(i+1)$th layer to jump into this hole. We will study the time it takes to nucleate a critical cluster of i_b atoms, and we will investigate how this nucleation time depends on the cluster location, i.e., the portion of the cluster is in the ring (i_R atoms) and the portion on the terrace [(i_b-i_R) atoms in the hole].

The total number $N(\rho_b,\rho_r,S)$ of possible distributions of atoms either within the ring or within the hole for a given pair (ρ_b,ρ_r) is

$$N(\rho_b,\rho_r,S)=\begin{pmatrix}6R\\\rho_b6R\end{pmatrix}\begin{pmatrix}S\\\rho_rS\end{pmatrix}$$

$$=\frac{(6R)!}{(\rho_b6R)!\,[6R(1-\rho_b)]!}\frac{S!}{(\rho_rS)!\,[S(1-\rho_r)]!}.$$

(11)

In the case of maximum S the expression above becomes

$$N(\rho_b,\rho_r,S)=\begin{pmatrix}6R\\\rho_b6R\end{pmatrix}\begin{bmatrix}3(R-1)R\\\rho_r3(R-1)R\end{bmatrix}$$

$$=\frac{(6R)!}{(\rho_b6R)!\,[6R(1-\rho_b)]!}$$

$$\times\frac{[3R(R-1)]!}{[\rho_r3R(R-1)]!\,[3R(R-1)(1-\rho_r)]!}.$$

(12)

Next we calculate the number of possible ways to distribute the i_b atoms which form the critical size cluster, (i_b,i_R), with i_R atoms in the ring and (i_b-i_R) outside is

$$N(i_b,i_R)=\begin{pmatrix}6R-i_R\\\rho_b6R-i_R\end{pmatrix}6R\begin{bmatrix}S-(i_b-i_R)\\\rho_rS-(i_b-i_R)\end{bmatrix}$$

$$=\frac{(6R-i_R)!}{(\rho_b6R-i_R)!\,[6R(1-\rho_b)-i_R]!}$$

$$\times\frac{[S-(i_b-i_R)]!}{[\rho_rS-(i_b-i_R)]!\,[S(1-\rho_r)-(i_b-i_R)]!},$$

(13)

for the maximum S the expression becomes

$$N(i_b,i_R)=\begin{pmatrix}6R-i_R\\\rho_b6R-i_R\end{pmatrix}6R\begin{pmatrix}3(R-1)R-(i_b-i_R)\\\rho_r3(R-1)R-(i_b-i_R)\end{pmatrix}.$$

(14)

For fixed i_b it is clear that these expressions show a strong dependence of $N(i_b,i_R)$ on the densities ρ_b, ρ_r. All configurations with the number of atoms in the ring $6R\rho_b$ and $S\rho_r$ in the hole can be realized with equal probability. The diffusion

TABLE II. The time of nucleation t_n as a function of i_R for two sets of values of the densities ρ_b shown according to Eq. (15). n_b is the number of atoms in the ring $\mathfrak{R}$, and ρ_r is calculated from the detailed balance condition Eq. (17); n_r is the number of atoms on top and outside the ring.

ρ_b	n_b	ρ_r	n_r	i_R	t_n [τ_0]
				8	5.5×10^{-2}
				7	15
0.76	136	0.0014	3	5	1.75×10^6
				3	5×10^{11}
				1	7.5×10^{17}
				8	2×10^{-2}
				7	3.25
0.85	153	0.0025	7	5	1.2×10^5
				3	6.75×10^9
				1	6×10^{14}

of atoms on surface sets the time scale how the system realizes these different configurations and therefore how often it can find the "correct" configurations with i_b atoms connected and therefore nucleate the next layer. During one time unit $(\rho_b 6R + \rho_r S)P_t$ new configurations are explored (where P_t has been defined in Sec. II and is the probability for an atom on the terrace to move to a neighboring position). During one time unit all atoms on the surface on average have a chance to diffuse. (We neglect that some atoms cannot move because the neighboring sites are blocked since nucleation is expected to happen at low density when blocking effects should be minimal.) The time for i_b atoms to become connected is the minimum time needed for nucleation. Since atoms within the ring have the same energy barrier for diffusion azimuthally as atoms on the terrace, P_t is independent of the position of the atoms. The jumps from the ring to the hole which have higher barrier E_b are neglected because these jumps have small probability and because of detailed balance they result in a higher density within the ring ρ_b vs within the hole ρ_r.

The rate of change of the configurations is given by $(\rho_b 6R + \rho_r S)P_t$. Within the time defined by the ratio of the total number of configurations to be explored and the rate of exploring them, eventually a configuration which includes a critical cluster will be realized. (It is assumed that the configurations with a critical size cluster can be found with equal probability starting from any initial configuration.) The mean time of the realization of configuration with critical cluster is therefore given by

$$t_n(i_b, i_R) = \frac{N(\rho_b, \rho_r, S)}{N(i_b, i_R)(\rho_b 6R + \rho_r S)P_t},$$

$$S \leq 3(R-1)R. \qquad (15)$$

From this definition of the nucleation time it is easy to see that the minimum nucleation is when $i_R = i_b$; i.e., the whole critical cluster is inside the ring.

Atoms on the ring diffuse around out of the ring or within, meet on the ring, and form clusters. The key difference is the large barrier $E_b = 0.17$ eV to diffuse out of the ring. Diffus-

ing azimuthally within the ring the barrier is $E_t = 0.05$ eV much smaller than E_b. This corresponds to a rate of diffusing within the one lattice constant ring at 180 K 2.3×10^3 times more than diffusing out of the ring. This means as the atoms are coming from the wetting layer they have a better chance to nucleate $i_c = 8$ atoms within the one lattice constant ring than to diffuse out of the ring into the terrace and form an 8 atom compact cluster with part in and part out of the ring. The large anisotropy $E_b \gg E_t$ is seen in the experiment and the use of one lattice constant ring was to be consistent with the earlier simulations (Ref. 15). In reality the ring width is bigger than one lattice constant but because the island radius used in Ref. 15 was 30 lattice constants, the ring width was limited to one lattice constant. This is also seen in Table II, where the nucleation time t_n is calculated according to Eq. (15) for two arbitrary values of the density ρ_b and for maximal $S = 2790$ when $R = 30$.

The time t_n defined in Eq. (15) should be compared to the time t_c to fill the island top, which was shown before Eq. (10) to have as the lower limit $S/J_{\text{WL-}\mathfrak{R}}$. Using the rate $J_{\text{WL-}\mathfrak{R}} = 6Rn_p \exp(-\frac{E_{\text{WL}\to t}}{kT})(1-\rho_b) = 9 \times 10^{-5}[\tau_0]^{-1}$ for atoms to move from the wetting layer to the island top, we obtain for $R = 30$ (or $S = 2790$), $T = 180$ K, $E_{\text{WL}\to t} = 0.25$ eV, $n_p = 5$ the time to complete a single layer (either on the sixth layer or seventh layer) is $t_c \approx S/J_{\text{WL-}\mathfrak{R}} = 3.1 \times 10^7[\tau_0]$.

The filling of the different layers requires different amount of Pb depending how much the layer below is complete. If we study the nucleation on top of the fifth layer (deposited atoms in sixth layer) then only one layer ($N = 2790$ positions) needs to be filled because there is no hole for atoms to descend into. For the eighth layer the ring should be filled by atoms in the sixth and seventh layer since both layers have holes. The time to complete the eighth layer requires the completion of both the sixth and seventh layers and is given by $t_c = 2S/J_{\text{WL-}\mathfrak{R}} = 6.2 \times 10^7[\tau_0]$; these values are in Table III.

We calculate the nucleation time t_n as a function of the atom density in the ring ρ_b for critical clusters which are fully contained within the ring (with $i_R = i_b$). t_n strongly depends on ρ_b as seen from the factorial terms in Eqs. (11)–(13). If we consider that the time of nucleation t_n should

TABLE III. Critical parameters for nucleation on different layers for $T = 180$ K and the barriers listed in Table I. The value of the critical density in the ring $\mathfrak{R}$ $\rho_b^c = 0.1$.

Parameter	Layer		
	6	7	8
t_c $[\tau_0]$	3.1×10^7	3.1×10^7 (monolayer), 6.15×10^7 (bilayer)	6.15×10^7
Time to the critical density $\rho_b^c = 0.1$	ρ_b^c reached at $t = 2 \times 10^5$	ρ_b^c reached at $t = 2 \times 10^5$	Never, $\rho_{max} = 0.074$
Nucleation	Yes	Yes	No

be less than the time t_c of the filling of top layer, we can estimate critical density of atoms on the ring as $\rho_b^c \approx 0.092[t_n(0.1) = 6.2 \times 10^7 \tau_0]$ for bilayer growth and $0.0975[t_n(0.1) = 3.1 \times 10^7 \tau_0]$ for the monolayer filling.

In Fig. 4 the dependence of the nucleation time on density ρ_b is presented from Eq. (15). This dependence is very strong as seen in Eq. (15) because for higher values of ρ_b the creation of critical cluster is very probable. For lower values of density ρ_b such creation is practically impossible and the critical cluster is never formed. The dependence of t_n on temperature T is determined by factor P_t, which is independent of density and only scales time. Therefore we focus our consideration only on part of the curve in Fig. 4 which is close to the time centered around 5×10^7 in units of τ_0 defined earlier (for the parameters used in Ref. 14).

We can also conclude that the nucleation starts on the ring, not in the hole. Because of detailed balance the term $\rho_r S$ is small, and t_n increases with decreasing i_R. For the case $i_b = i_R$, where the time of nucleation is minimal, the dependency $t_n(\rho_b)$ is the same for all layers, because it does not depend on S—it cancels in $N(\rho_b, \rho_r, S)/N(i_b, i_R)$.

Before we analyze the possibility of nucleation of critical cluster on layers 7 and 8, we discuss next the analysis to derive the time dependence of the densities ρ_b of atoms as a function of time, which plays an important role to see
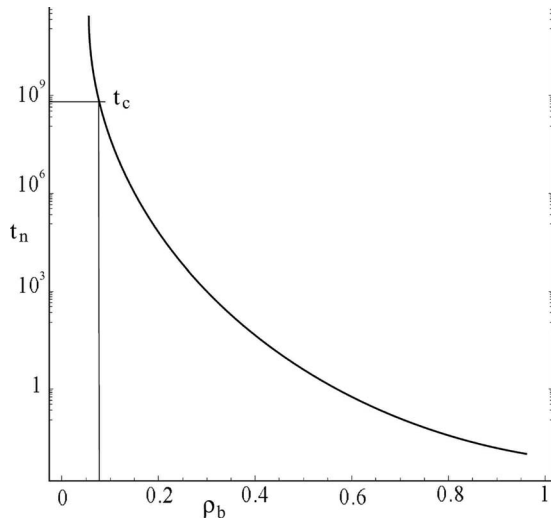

FIG. 4. t_n as a function of ρ_b layer defined by Eq. (15). This is a measure of the nucleation time of the critical cluster for the specific case $i_b = 8$, $i_R = 8$. The temperature is $T = 180$ K, the critical density $\rho_c \approx 0.1$, and the rest of the parameters are listed in Table I.

whether the critical density defined in Fig. 4 is attained before or after the completion time t_c.

VI. NUCLEATION IN THE SIXTH LAYER

In this paper we study the growth of islands from the fifth layer to seventh layer. It implies that initially the island is five layers high so the fifth layer is completely full.

If we analyze the nucleation in the sixth layer, one of the conditions is that the fifth layer is already filled so there is no central hole for atoms to descend into. In this case the area of the central region S outside the ring is maximal. There is no flux from the sixth to the fifth layer $J_{-1} = 0$.

The density of atoms in the ring $\mathfrak{R}$ can be determined from the balance of fluxes. Incoming fluxes adding particles to the density ρ_b are as follows: $J_{\text{WL-}\mathfrak{R}}$ from the wetting layer (WL) and $J_{t\text{-}\mathfrak{R}}$ from the central area back to the ring rest. Outgoing fluxes reducing ρ_b are $J_{\text{WL}}^{\text{tot}}$ (atoms jumping from the ring back to the wetting layer and $J_{\mathfrak{R}\text{-}t}$ atoms exiting the ring toward the central area):

$$\frac{d\rho_b}{dt} = \frac{1}{6R}(J_{\text{WL-}\mathfrak{R}} + J_{t\text{-}\mathfrak{R}} - J_{\text{WL}}^{\text{tot}} - J_{\mathfrak{R}\text{-}t})$$

$$= -\rho_b\left[n_p \exp\left(-\frac{E_{\text{WL}\to t}}{kT}\right) + \frac{1}{3}\frac{R-1}{R}\rho_r \exp\left(-\frac{E_t}{kT}\right)\right.$$

$$+ \frac{1}{3}(1-\rho_r)\exp\left(-\frac{E_b}{kT}\right)$$

$$\left.+ \frac{1}{3}\exp\left(-\frac{E_{t\to\text{WL}}}{kT}\right)\right] + n_p \exp\left(-\frac{E_{\text{WL}\to t}}{kT}\right)$$

$$+ \frac{1}{3}\frac{R-1}{R}\rho_r \exp\left(-\frac{E_t}{kT}\right). \quad (16)$$

These fluxes are normalized to the island perimeter $6R$ because we are considering the net flux in and out of the ring per unit length of the perimeter.

Since the energy barrier E_t for the diffusion within a layer is much smaller than the energy barriers $E_{\text{WL}\to t}$, $E_{t\to\text{WL}}$, E_b, we assume in addition that detailed balance holds at the inside ring perimeter between the local density of atoms entering vs leaving the ring:

$$\rho_b(1-\rho_r)\exp\left(-\frac{E_b}{kT}\right) = \rho_r(1-\rho_b)\exp\left(-\frac{E_t}{kT}\right). \quad (17)$$

This condition can be translated into a relation between $\rho_b \gg \rho_r$; i.e., the large barrier for atoms to exit the ring implies

much higher density inside than outside the ring. For the barriers used in the Table I, the corresponding Boltzmann factor is very low $[\exp(-\frac{E_b - E_t}{kT}) \approx 8.7 \times 10^{-4}]$, which can be used to deduce approximately

$$\rho_r = \exp\left(-\frac{E_b - E_t}{kT}\right) \frac{\rho_b}{1 - \rho_b\left[1 - \exp\left(-\frac{E_b - E_t}{kT}\right)\right]}$$

$$\approx \exp\left(-\frac{E_b - E_t}{kT}\right) \frac{\rho_b}{1 - \rho_b}. \quad (18)$$

Although this was calculated for one temperature $T = 180$ K it will be valid for other temperatures because of the larger barrier to exit the ring than to diffuse on the terrace $E_b > E_t$. For the beginning of the transition to the next layer ρ_b is also small which will also imply $(1 - \rho_b) \approx 1$ and therefore the density just inside the central area will be more than 3 orders of magnitude lower than the density in the ring $\rho_r / \rho_b \approx 8.7 \times 10^{-4}$. Additionally not only is the density ρ_r very low, but detailed balance also suggests that the two fluxes $J_{t-\mathfrak{R}}$ and $J_{\mathfrak{R}-t}$ are equal and Eq. (16) reduces to

$$\frac{d\rho_b}{dt} = \frac{1}{6R}(J_{\mathrm{WL}-\mathfrak{R}} - J_{\mathrm{WL}}^{\mathrm{tot}}) = -\rho_b\left[n_p \exp\left(-\frac{E_{\mathrm{WL}\to t}}{kT}\right)\right.$$

$$\left. + \frac{1}{3}\exp\left(-\frac{E_{t\to\mathrm{WL}}}{kT}\right)\right] + n_p \exp\left(-\frac{E_{\mathrm{WL}\to t}}{kT}\right) = A_6\rho_b + B,$$

$$(19)$$

where

$$A_6 = -\frac{1}{3}\exp\left(-\frac{E_{t\to\mathrm{WL}}}{kT}\right) - n_p \exp\left(-\frac{E_{\mathrm{WL}\to t}}{kT}\right) \approx -9$$

$$\times 10^{-7}[\tau_0]^{-1},$$

$$B = n_p \exp\left(-\frac{E_{\mathrm{WL}\to t}}{kT}\right) \approx 5 \times 10^{-7}[\tau_0]^{-1}. \quad (20)$$

The interpretation of this equation is that for very long times the density in the ring is fully determined by the balance of atoms climbing the edge barrier and the ones returning back to the wetting layer. The atoms at the inside radius of the ring play no role because the time constant of diffusion on the island top is much faster. The solution of Eq. (19) under initial condition $\rho_b = 0$ at $t = 0$ is

$$\rho_b = \frac{B}{A_6}[\exp(A_6 t) - 1]. \quad (21)$$

This means that the limit of ρ_b is $B/A_6 = 0.557$. The value lower than 1 is due to the blocking factor $(1 - \rho_b)$. Without this effect, the density on the ring can reach 1. Even with this limit it is much higher than critical density for nucleation.

Figure 5(a) shows the time dependence of $\rho_b(t)$ for the sixth layer. The final density B/A_6 is well above the critical density ($\rho_b^c = 0.1$ ML which as shown previously is independent of the layer because it is simply determined by the terrace diffusion on the top). This is attained in $2 \times 10^5 \, \tau_0$ which is a small fraction of the time to complete the layer

$\tau_c = 3.1 \times 10^7$. According to this time dependence, ρ_b increases with time [see Fig. 5(a)] quickly, and although from Eq. (17) ρ_r also increases, it remains very small until ρ_b reaches its maximum value B/A_6, well below the critical density on the central area which is higher than ρ_b^c (see Table II). It is due to the small value of ratio $\exp(-E_b/kT)/\exp(-E_t/kT) \approx 4.37 \times 10^{-4}$.

VII. NUCLEATION OF THE SEVENTH LAYER

We investigate the nucleation on the seventh layer with two differences from the barriers controlling nucleation on the sixth layer discussed above: atoms on the seventh layer can escape to the sixth layer from the inside perimeter of the growing ring because transitions from $7 \to 6$ have a finite energy barrier and atoms have lower probability to move to the wetting layer because the barrier is higher $E_{t\to\mathrm{WL}}$. The two changes have different contributions to the density in the ring and based on the barriers listed in Table I, the second effect is more important. The island density in the ring exceeds again the critical density ρ_b^c quickly within the time $2 \times 10^5 \, \tau_0$, which is a small fraction of the completion time t_c. We can use the same Eq. (19) for the time evolution of the density ρ_b,

$$\frac{\partial \rho_b}{\partial t} = \frac{1}{6R}[J_{\mathrm{WL}-\mathfrak{R}} + J_{t-\mathfrak{R}} - J_{\mathrm{WL}}^{\mathrm{tot}} - J_{\mathfrak{R}-t}]. \quad (22)$$

The difference with nucleation on the sixth layer is in the flux $J_{-1} = 2r\rho_r \exp(-\frac{E_{7\to6}}{kT})$. Here we assume the barrier for the $7 \to 6$ jumps, $E_{7\to6} = 0.2$ eV, and we must estimate the role of the increasing ρ_r because of this finite flux on the dependence of the density ρ_b in the ring. J_{-1} also depends linearly on the radius of central vacancy r and because r decreases with time, it follows that the number of sites for atoms to move to the lower level also is reduced. We assume as before detailed balance because both E_t and $E_{7\to6}$ barriers are lower than the barrier to escape from the ring to the central area:

$$J_{\mathfrak{R}-t} = J_{t-\mathfrak{R}} + J_{-1},$$

$$2R\rho_b(1-\rho_r)\exp\left(-\frac{E_b}{kT}\right) = 2(R-1)\rho_r(1-\rho_b)\exp\left(-\frac{E_t}{kT}\right)$$

$$+ 6r\rho_r \exp\left(-\frac{E_{7\to6}}{kT}\right). \quad (23)$$

But despite the additional flux J_{-1} the flux to the seventh layer ring area from outside the ring $2(R-1)\rho_r(1-\rho_b)\exp(-\frac{E_t}{kT})$ is much larger than the flux from this area down to the sixth layer $6r\rho_r \exp(-\frac{E_{7\to6}}{kT})$. (It is approximately $0.33 \times \exp[-(E_t - E_{7\to6})/kT] \approx 5.3 \times 10^3$, which implies that we can neglect the flux J_{-1} and we obtain the same equation as in the case of sixth layer.)

More importantly, because the seventh layer is a stable height, the barrier for the jumps of atoms from perimeter to wetting layer $E_{t\to\mathrm{WL}}$ is higher than the corresponding barrier on sixth layer. This implies that the long time value ρ_b that can be attained on the seventh layer as seen from Eq. (19) applied to the seventh layer B/A_7 is higher. Using the barrier
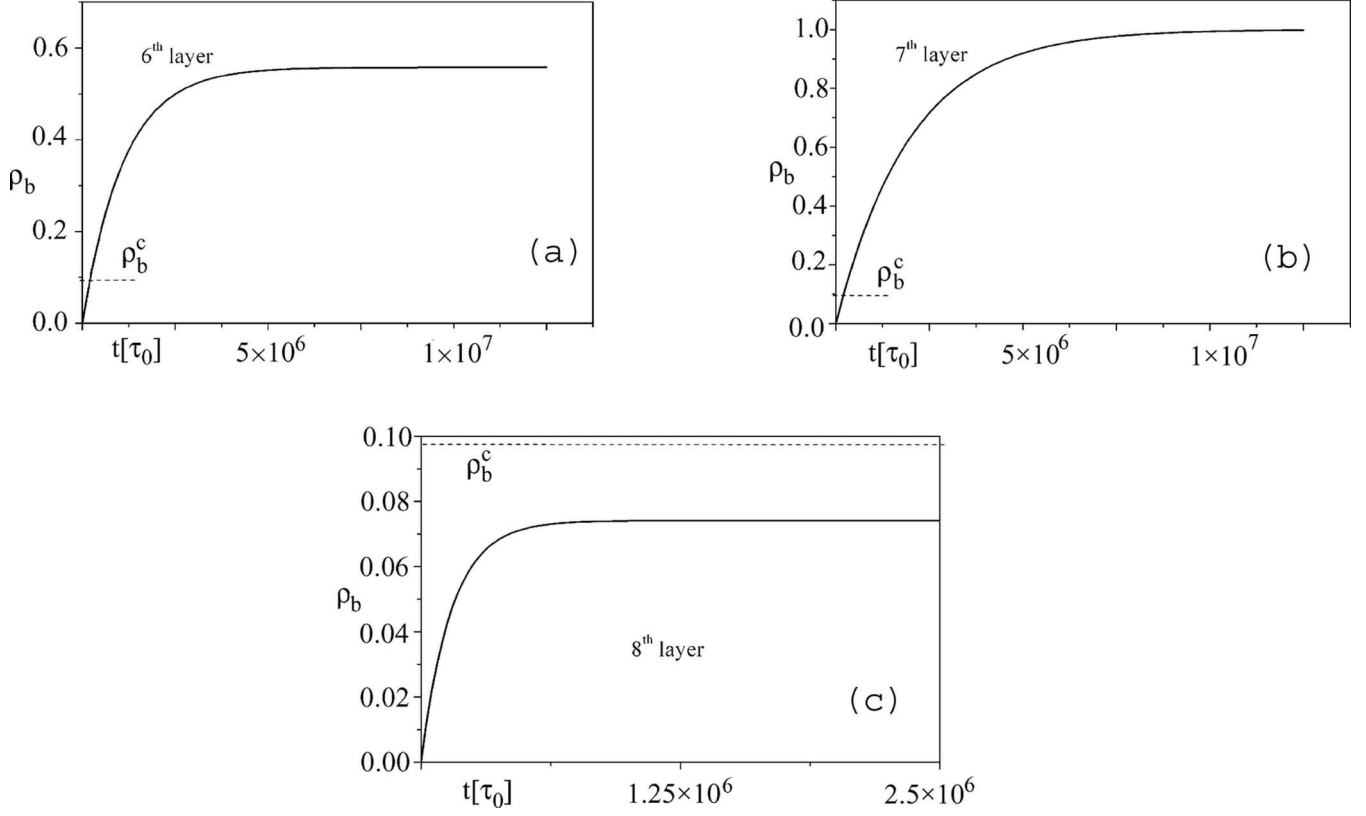
FIG. 5. $\rho_b(t)$ vs t time dependence of the density of atoms in ring $\mathfrak{R}$ for (a) the sixth layer, (b) for the seventh layer, and (c) for the eighth layer. The dashed line marks the critical density, i.e., the minimum density so nucleation in the layer can occur. For the sixth and seventh layers this density is attained relatively quickly but for the eighth layer is never attained because the flux of atoms back to the wetting layer and to the lower inside layers is higher. These results are for 180 K and the barriers of Table I but the conclusions are more generally applicable within the temperature range $140 < T < 220$ K consistent with the experiment.

from Table I $E_{t\rightarrow\text{WL}} = 0.32$ eV we have numerical values,

$$\rho_b = \frac{B}{A_7}[\exp(A_7 t) - 1],$$

$$A_7 = -\frac{1}{3}\exp\left(-\frac{E_{t\rightarrow\text{WL}}}{kT}\right) - n_p \exp\left(-\frac{E_{\text{WL}\rightarrow t}}{kT}\right) \approx -5 \times 10^{-7},$$

(24)

and we see that in this case the density increases as in the case of sixth layer, but the limit is $\rho_b = 1$. The critical density is reached at time approximately $2 \times 10^5 \, \tau_0$ as seen in Fig. 5(b) which is the same as the time to nucleate within the ring of the sixth layer. The more important conclusion is, as far as the nucleation and the existence of a bilayer ring is concerned, that nucleation is possible on both the sixth and seventh layers. It means the system of atoms on the seventh layer has *enough* time to nucleate on the perimeter as seen in Fig. 5(b).

VIII. ABSENCE OF NUCLEATION ON LAYER 8

Finally we investigate the puzzling result why there is no nucleation on the eighth layer although the barrier $E_{t\rightarrow\text{WL}}$ is the same on the sixth and eighth layers. First the flux from

the top to the wetting layer is higher than from the top of the seventh layer because the barrier $E_{t\rightarrow\text{WL}}$ is lower. In addition the energy barrier for jumps down to seventh or sixth layer is zero. Both these effects result in removing atoms from the ring, either back to the wetting layer or to the lower layers on the inside perimeter of the ring, so they reduce ρ_b on the eighth layer below ρ_b^c and prohibit nucleation. In the case of the eighth layer, these effects and the fast diffusion on the terrace imply that all atoms, which leave the ring at the inside perimeter, fall toward the central vacant area and thus $J_{-1} = J_{\mathfrak{R}\text{-}t}$ and $J_{t\text{-}\mathfrak{R}} = 0$. The time evolution of ρ_b in the form includes only three terms since no back hops from the central area to the ring are possible,

$$\frac{\partial \rho_b}{\partial t} = J_{\text{WL-}\mathfrak{R}} - J_{\mathfrak{R}\text{-WL}} - J_{\mathfrak{R}\text{-}t} = \left[n_p(1 - \rho_b)\exp\left(-\frac{E_{\text{WL}\rightarrow t}}{kT}\right) \right.$$

$$\left. - \rho_b \frac{1}{3}\exp\left(-\frac{E_{t\rightarrow\text{WL}}}{kT}\right) - \rho_b \frac{1}{3}\exp\left(-\frac{E_b}{kT}\right) \right]. \quad (25)$$

The right-hand side of this equation is

$$n_p(1 - \rho_b)\exp\left(-\frac{E_{\text{WL}\rightarrow t}}{kT}\right) - \rho_b \frac{1}{3}\exp\left(-\frac{E_{t\rightarrow\text{WL}}}{kT}\right)$$

$$- \rho_b \frac{1}{3}\exp\left(-\frac{E_b}{kT}\right) = A_8\rho_b + B,$$

$$A_8 = -4.26 \times 10^{-5} [\tau_0]^{-1}. \tag{26}$$

The solution of this equation is

$$\rho_b = \frac{B}{A_8}[\exp(A_8 t) - 1]. \tag{27}$$

Because one of the fluxes is zero A_8 takes a larger value (in magnitude) which will imply the density at very long times will be B/A_8 lower. The value of A_8 is approximately 2 orders of magnitude than for the sixth and seventh layers so it reduces the asymptotic value of ρ_b and therefore the probability to nucleate on the eighth layer. Limits of the density is $\rho_b = 0.074$ less than the critical value. If we calculate the time of nucleation in the case of maximal $\rho_b = 0.074$, it is $t_n(0.074) = 8 \times 10^8 \ \tau_0$. This time is ten times higher than the time of filling of bilayer $t_c = 6.15 \times 10^7 \ \tau_0$. In this case the ρ_b^c is not realized. The result is shown in Fig. 5(c) which demonstrates that nucleation is not possible. Our results are summarized in Table III.

The calculations were shown for one temperature $T = 180$ K corresponding to the experiment of Ref. 16. They depend on the differences between the barriers of Table I that define the two parameters B_i and A_i (with i as the island height) in Eqs. (21), (24), and (27) that determine the long time island density ρ_b. All conclusions about which layer will support nucleation remain the same within the temperature range 140–220 K. Only for $T > 200$ K the asymptotic value of $\rho_b = \rho_b^c$ exceeds the critical value so nucleation at the eighth layer is allowed. As seen in Fig. 1 of Ref. 15 it is possible to nucleate on top of the bilayer ring when growth is at 240 K but the increments are either five layers (so the ring height measured from the wetting layer is nine layers) or seven layers (so the total ring height measured from the wetting layer is 11 layers). This indicates that the dependence of the barrier to fall from island top to the wetting layer on island height for stable layers is lower than for unstable layers and is still applicable even at larger height islands.

IX. CONCLUSIONS

In this paper we introduce an analytic model of the nucleation on the perimeter of islands during self-assembled growth of the uniform height Pb islands on Si(111) surface to increase the transparency and predictability of the Monte Carlo simulation results. This model explains the interesting feature of the formation of bilayer ring at the edge of islands during the growth from stable to stable heights. One question is central to the description of such growth mode: why is the nucleation through a bilayer ring and why is there no nucleation observed on the third layer if atoms are arriving on the third layer from the wetting layer? In the model we include the flux of atoms from the wetting layer to the top layer and the opposite flux of atoms from the top to the incomplete lower layers on the inside perimeter or back to the wetting layer from the outside perimeter of the ring. In addition the preference of atoms to nucleate on the perimeter of the top layer is defined by a ring of one lattice constant width. The fluxes of atoms depend on the layer (if it is stable or unstable) and reflect the role played by QSE. Because the barrier to go back to the wetting layer from the island top depends on whether the island is stable or unstable, unstable islands have lower barriers than stable islands; i.e., atoms within unstable heights fall easier than atoms within stable heights as concluded from the simulations of Refs. 14 and 15.

Our results can be applied also for other systems where QSE play a role and results in similar uniform height growth modes. In addition if these key barriers are known (from kinetic measurements or can be calculated from first-principles calculations) then it would be possible to use the model we formulated in this work to be able to identify the optimal temperature and coverage window where the sharpest island height distribution can be obtained, i.e., when nucleation of unstable on stable heights is suppressed.

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