

Kinetics of second layer nucleation with permeable steps



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ABSTRACT

The critical island size for second layer nucleation is a basic parameter which determines the mechanism of growth and in turn the morphology of the growing surface. When it is larger than the mean distance between the islands layer-by-layer growth takes place. In the opposite case 3D mounds are formed. The critical island size for second layer nucleation is calculated by taking into account the ability of adatoms to cross the step surrounding the island without joining the kink sites (the effect of the step permeability). We have found that the mean field approximation fails to give solution in the case of permeable steps. By making use of the probabilistic approach for derivation of the nucleation frequency on top of the first layer island we show that the critical size decreases with increasing degree of permeability. This is due to the contribution of atoms from the area surrounding the island which is especially important at the early stage of deposition where the average size of the first layer islands is small. The decrease of the critical size is steeper at higher temperatures or smaller densities of the first layer nuclei and in turn larger areas around the islands which feed the latter. The rate of growth of the first level island remains unaffected by the step permeability as the adatoms always join the step after several (or many) attempts under the condition of complete condensation. An important conclusion is that the step permeability could give rise to a transition from planar to 3D growth.

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1. Introduction

The morphology of a growing crystal surface is of crucial importance for device applications. In most cases a smooth surface is highly desirable. The latter is a result of a planar, two-dimensional (2D) growth. However, in many cases, pyramids of growth or mounds which represent stacks of 2D monolayer islands are formed [1]. The formation of mounds is due to the existence of a kinetic barrier for down-step diffusion known as Ehrlich–Schwoebel (ES) barrier [2,3]. The latter gives rise to an effective uphill current and a higher probability of nucleation on the top island, and in turn to unstable growth, as has theoretically been predicted by Villain [4]. It is thus clear that the nucleation on the topmost island of the stack, the island-on-island or very often second-layer nucleation as is known in the literature, plays a key role in the mechanism of growth and in turn on the morphology of the growing surface [5].

A nucleus of the second layer is formed when the adatom concentration on top of the first layer island reaches a critical value and thus a critical supersaturation. It was Chernov who found that this concentration is ultimately connected with a critical size of the first layer island [6]. The adatom concentration has a profile of a dome on top of the island surface and the maximum in the middle increases with the square of

the island's radius. When the critical island size for second layer nucleation is larger than the mean spacing between the islands the first layer islands cover completely the surface before the nuclei of the second layer are formed on top and layer-by-layer growth takes place. In the opposite case, the nuclei of the second layer are formed before the complete coverage of the growing surface by the first layer islands and the formation of mounds or 3D growth possible. The second layer nucleation thus plays a crucial role in determining the morphology of the growing surface.

Stoyanov and Markov calculated the critical radius in diffusion limited growth by using a mean-field approximation to explain the transition from planar growth to three-dimensional islanding in heteroepitaxy [7,8]. Tersoff calculated later the critical island's radius in a kinetic regime of growth assuming the existence of the ES barrier to down-step diffusion again within the framework of the mean-field approximation [9]. The result in Ref. [9] has been used for calculation of the ES barrier in a series of papers [10–14]. Rottler and Maass [15] showed that a kinetic Monte Carlo simulation disagreed with the mean field approach of [9] for nuclei consisting of 1 and 2 atoms and concluded that in this case the stochastic character of the processes involved plays a crucial role in determining the nucleation rate on top of the island. Krug, Polity and Michely developed further the probabilistic treatment of Rottler and Maass and applied the results to the formation of mounds or wedding cakes. They found that the probability P_{i+1} a $i+1$ st atom to arrive on the surface while i atoms are still present on it is given by $(\tau/\Delta t)^i/i!$,

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where τ is the time of residence of the atoms on the island, and Δt is the average time period of arrival of atoms [16]. Comparison with the mean-field approach showed that the latter overestimates the nucleation frequency of the second layer by a factor $l_{ES}/R \gg 1$ where $l_{ES} = \exp(E_{ES}/kT)$ is the Ehrlich–Schwoebel length, E_{ES} is the Ehrlich–Schwoebel barrier to down-step diffusion, and R is the island's radius (all lengths are relative to the atom spacing a). They concluded that in the case of $i = 1$ the island's surface is empty most of the time, and sometimes it is populated by a single atom, and very rarely it happens that a second atom arrives during that time. Once the two atoms are simultaneously present on the island a nucleus is formed with a probability close to unity (the lonely adatom model). Further considerations of Heinrichs, Rottler and Maass showed convincingly that the mean-field approach is applicable for critical nuclei consisting of more than 3 atoms when the number of atoms on the first layer island is sufficiently greater than the number of atoms in the critical nucleus [17]. If the nucleus is smaller (one or two atoms) the random character of the processes involved becomes significant. The scaling regimes for second layer nucleation were summarized and generalized in Refs. [17] and [18]. For a recent review see Ref. [19].

The aim of the present paper is to study the effect of the step permeability introduced by Ozdemir and Zangwill [20] on the critical island size for second layer nucleation and thus on the roughening of the crystal surface. For doing that we make use of both the mean-field approximation and the stochastic approach to calculate the nucleation frequency on the top surface of the island. We make use of the terrace-edge-kink model considered in detail by Filimonov and Hervieu [21]. The latter includes diffusion of adatoms to the step, diffusion along the step edge and either incorporation into kink site or jump to the neighboring terrace (step permeability). It thus gives the interrelation between the kinetic coefficients for the incorporation of atoms into kink sites and the coefficient of the step permeability. We show that the step permeability leads to an increase of the frequency of the second layer nucleation and in turn to a decrease of the critical island size. The latter makes possible the transition from layer-by-layer to multilayer or 3D growth and rough crystal surface.

2. Critical island size

In analogy with the definition of Chernov we write the critical size, R_c , for second layer nucleation in integral form [22]

$$\int_0^{R_c} \frac{\Omega(R)}{v(R)} dR = 1 \quad (1)$$

where $\Omega(R)$ is the nucleation frequency defined as

$$\Omega(R) = \int_0^R J[n_1(r)] 2\pi r dr \quad (2)$$

and

$$v(R) = \frac{dR}{dt} \quad (3)$$

is the rate of growth of the 2D island with radius R , and $n_1(r)$ is the profile of the adatom concentration on top of the island (level 1).

The nucleation rate is given by [23]

$$J(r) = C n_1(r)^{i+1} \quad (4)$$

where i is the number of atoms in the critical nucleus, and

$$C = \alpha^* D N_0 \exp(E^*/kT) \quad (5)$$

where α^* is the number of ways an atom can join the critical nucleus to produce a stable cluster, $D = \nu \exp(-E_{sd}/kT)$ is the surface diffusion

(hopping) frequency, ν is the attempt frequency, and E_{sd} is the respective activation barrier. $N_0 = a^{-2}$ is the atom surface density, and E^* is the binding energy of the nucleus. The adatom concentration is relative to the surface atom density N_0 . For $i = 1$, $C = \alpha^* D N_0$.

2.1. Growth rate of first layer island

We calculate first the growth rate, $v(R)$, of the first layer island (see Fig. 1). In the calculations below all lengths are in units of the atom spacing a and the concentrations are relative to the adatom surface density $N_0 = 1/a^2$. We assume that the island belongs to an ensemble of regularly spaced equidistant islands with density N_s and with one and the same size. We thus use the mean field approximation. The islands grow at the expense of atoms diffusing to their edges on the substrate (level 0) and on top of the islands (level 1).

The adatom concentration on level 0 (around the island) is found from the solution of the diffusion equation in polar coordinates on the ring enclosed by R and L

$$n_0(r) = A - \frac{F}{4D} r^2 + B \ln(r) \quad (6)$$

where A and B are the constants which can be found by the boundary conditions $n(R) = \bar{n}_0$, \bar{n}_0 being the adatom concentrations in the near proximity of the ascending step, and $dn_0(r)/dr = 0$ in the middle between neighboring islands ($r = L$). Then

$$n_0(r) = \bar{n}_0 + \frac{F}{4D} (R^2 - r^2) + \frac{FL^2}{2D} \ln\left(\frac{r}{R}\right) \quad (7)$$

where $\pi L^2 = 1/N_s$ is the area around the island which feeds the latter, and F is the atom arrival frequency.

The adatom concentration on level 1 (on top of the island) is [7]

$$n_1(r) = \bar{n}_1 + \frac{F}{4D} (R^2 - r^2) \quad (8)$$

where \bar{n}_1 is the adatom concentration in the proximity of the descending step which encloses the island.

The concentrations \bar{n}_0 and \bar{n}_1 are determined from the boundary conditions [20]

$$\begin{aligned} D \frac{dn_0(r)}{dr} \Big|_{r=R} &= +K_0(\bar{n}_0 - n_{eq}) + K_p(\bar{n}_0 - \bar{n}_1) \\ D \frac{dn_1(r)}{dr} \Big|_{r=R} &= -K_1(\bar{n}_1 - n_{eq}) - K_p(\bar{n}_1 - \bar{n}_0) \end{aligned} \quad (9)$$

where K_i ($i = 0, 1$) are the kinetic coefficients for incorporation of atoms to kink sites from the above terrace (subscript 1) and from the substrate (subscript 0), K_p is the coefficient of the step permeability, and n_{eq} is the equilibrium adatom concentration.

From Eqs. (7), (8) and (9) one obtains

$$\begin{aligned} DMq \frac{1-\theta}{\sqrt{\theta}} &= K_0 X_0 + K_p(X_0 - X_1) \\ DMq \frac{\theta}{\sqrt{\theta}} &= K_1 X_1 + K_p(X_1 - X_0) \end{aligned} \quad (10)$$

where $X_i = \bar{n}_i - n_{eq}$, ($i = 0, 1$), $q = \sqrt{4\pi N_s}$, $M = F/(4\pi D N_s)$, and $\theta = \pi R^2 N_s = R^2/L^2$.

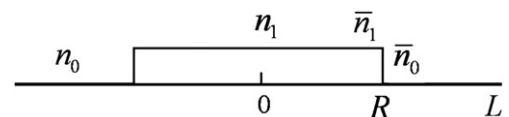


Fig. 1. Model for calculation of the growth rate $v(R)$ of the first layer island.

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