

## Critical terrace width for step flow growth: Effect of attachment-detachment asymmetry and step permeability

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A general expression for the critical terrace width for step flow growth accounting for both the step permeability and the asymmetric incorporation of atoms to ascending and descending steps is derived. It covers both cases of diffusion and attachment-detachment limited regimes of growth at high and low temperatures. It is found that when at least one of the excess step-edge barriers is equal to zero, only diffusion-limited behavior regime is allowed. The step permeability does not affect both limiting cases at high and low temperatures. Comparing the theory with experimental data for the Si(111)(7×7) surface leads to the conclusion that the nucleation process in the interval 700–850 K takes place in an attachment-detachment limited regime with critical nuclei consisting of one and three atoms below and above 750 K, respectively. The values of 0.525 and 0.535 eV have been estimated for the energy barriers for incorporation of adatoms into descending and ascending steps, respectively, thus demonstrating an inverse Ehrlich-Schwoebel effect [J. Chem. Phys. **44**, 1039 (1966)]. A binding energy of 1.65 eV for three-atom nucleus has been evaluated.

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Step flow and two-dimensional island nucleation and growth (2DNG) are two basic mechanisms of crystal growth.<sup>1</sup> A crystal grows exclusively by step flow when the temperature is sufficiently high so that the mean free path of the adatoms is longer than the mean terrace width. Decreasing temperature leads to a drastic decrease of the adatom diffusivity, which gives rise to island nucleation and growth on the terraces.<sup>1,2</sup> The interplay of surface diffusion and step attachment in growth produces an adatom concentration on terraces that generally increases with increasing spacing between step sinks. Consequently, a transition between step flow and 2DNG occurs as a function of terrace width. This transition is characterized by a quantity called the critical terrace width. Since growth involves a number of thermally activated processes, it is not surprising that the critical terrace width also displays an Arrhenius behavior as a function of deposition temperature.<sup>3–5</sup>

Island nucleation occurs with greatest probability at the points where the adatom concentration is the highest.<sup>6</sup> Therefore, a full understanding of the transition between step flow and 2DNG requires accurate knowledge of the adatom concentration profile on a terrace. The latter is determined during growth by several factors. This includes the relative importance of diffusion and step attachment, i.e., the rate-limiting step, which is characterized in opposite extremes as attachment-detachment limited (ADL) and diffusion-limited (DL) regimes. Asymmetry of the kinetic coefficients for step attachment from opposite sides of a step<sup>7</sup> and step permeability<sup>2,8</sup> are also known to affect the adatom concentration profile. Therefore, the critical terrace width should depend on the rate-limiting step, the kinetic coefficient asymmetry, and step permeability, and may be used, in principle, to determine these important microscopic details. Various models of the critical terrace width have been presented that loosely correspond to the different extremes of the rate-limiting step.<sup>4,9,10</sup> However, these models did not take into account the kinetic coefficient asymmetry or step permeabil-

ity. In this paper, we develop a more general model of the critical terrace width. This model, which is based on a solution for the adatom concentration for the case of complete condensation (complete absence of reevaporation), takes into account the kinetic coefficient asymmetry and step permeability and is applicable for rate-limiting kinetics that vary continuously between the ADL and DL extremes. This model is then applied to previously published data for the Si(111) × (7 × 7) surface.<sup>5</sup>

An important point should be clarified. In the case of incomplete condensation (adsorption-desorption equilibrium far from the steps) which is considered by Burton *et al.*,<sup>1</sup> the rate of step propagation depends on the rate-limiting step: attachment-detachment or surface diffusion. In the extreme of complete condensation, the rate of step advance is simply given by the atom arrival rate multiplied by the terrace width<sup>11</sup> and depends neither on the step attachment nor on the surface diffusion. It is the nucleus density  $N$  which depends on the rate-limiting step through the scaling exponent  $\chi$  in the expression for the nucleus density  $N \propto (D/F)^\chi$ , where  $D$  and  $F$  are the diffusion and atom arrival frequencies.<sup>6</sup>

We consider a train of equidistant straight steps. We solve the diffusion problem for the adatom concentration per site,  $n(x)$ , on a terrace with a width  $\lambda$  (in units of the adsorption sites separation  $a$ ) for the case of complete condensation,

$$D_s \frac{d^2 n(x)}{dx^2} + R = 0, \quad (1)$$

where  $R$  and  $D_s = a^2 \nu \exp(-E_{sd}/kT)$  are the atom arrival rate and the surface diffusion coefficient,  $\nu$  is the attempt frequency, and  $E_{sd}$  is the diffusion barrier on the terrace.

The solution reads<sup>12</sup>

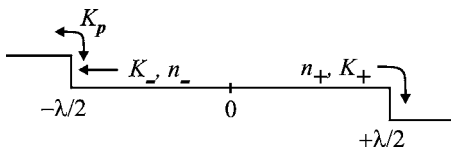


FIG. 1. The model for calculation of the adatom concentration as a function of the distance from the middle of the terrace, with the step kinetic coefficients and the respective adatom concentrations at the terrace boundaries.

$$n(x) = \frac{1}{2}(n_+ + n_-) + (n_+ - n_-) \frac{x}{\lambda} + \frac{R}{2D_s} \left( \frac{\lambda^2}{4} - x^2 \right), \quad (2)$$

where  $n_+$  and  $n_-$  are the adatom concentrations in the vicinity of the right-hand (descending) and left-hand (ascending) steps, respectively (see Fig. 1). The steps propagate from left to right. All quantities having dimension of a length are taken relative to the atomic diameter  $a$ .  $n_+$  and  $n_-$  can be found from the boundary conditions<sup>8</sup>

$$\left( \frac{dn}{dx} \right)_{\pm\lambda/2} = \mp K_{\pm}(n_{\pm} - n_{\text{eq}}) \mp K_p(n_{\pm} - n_{\mp}), \quad (3)$$

where  $n_{\text{eq}}$  is the equilibrium adatom concentration, and  $K_-$  and  $K_+$  are the kinetic coefficients for attachment to the ascending step at the left-hand side ( $x=-\lambda/2$ ) of the terrace and the descending step at the right-hand side ( $x=+\lambda/2$ ), respectively (Fig. 1). The middle of the terrace is taken as the origin,  $x=0$ .

The kinetic coefficients of the steps are of the form<sup>13</sup>

$$K_{\pm} = \left[ \exp\left(\frac{E_{\pm}}{kT}\right) - 1 \right]^{-1}, \quad (4)$$

where  $E_+$  and  $E_-$  are the respective energy barriers in excess of the barrier of surface diffusion. This form of the kinetic coefficients allows  $K_{\pm} \rightarrow \infty$  at  $E_{\pm}=0$ .<sup>13</sup>

The coefficient  $K_p$  accounts for the step permeability. Its definition is not straightforward. The step permeability has been considered as a phenomenon in which the atoms repeatedly join smooth (kink-free) parts of the step, diffuse along the edge, and leave the step without joining a kink site.<sup>14,15</sup> When atoms join kink sites, they become a part of the crystal lattice. It should be noted, however, that the irreversible attachment of an atom to the crystal lattice is not a result of a single event of joining a kink site but of many “trials and errors.”<sup>16</sup> An atom can repeatedly join a kink site and leave it to be adsorbed at a kink-free part of the step edge and after that to desorb from it and go back on the terrace. Thus, the atoms become, at least for a time, part of the crystal lattice. Following the above authors, we exclude such events from the definition of the step permeability. Then, the step permeability and the step impermeability should be considered as complementary phenomena in the overall attachment-detachment process.<sup>15,17</sup> However, assuming some degree of permeability in the particular case of complete condensation means that if an atom does not join a kink site at a particular step, it will do that at another step. If the permeability is zero, it will join a kink at the first step which it meets. Increasing the degree of permeability from zero onward, the

number of steps the atom will cross without joining a kink will increase. Some steps can be visited only once, some steps several times. However, at the end of its journey the atom will inevitably join a kink site. That is why we introduce formally the kinetic coefficient  $K_p$  as an independent parameter having the same form as  $K_{\pm}$ ,  $E_p$  being some effective energy barrier which includes in a complicated way the barriers of the processes involved (attachment to and detachment from smooth parts of the step, step-edge diffusion, etc.). The barrier  $E_p$  can vary from zero (complete permeability) to infinity (complete impermeability).

The adatom concentration then reads

$$n = n_{\text{eq}} + \frac{R\lambda^2}{2D_s} \left[ \frac{p}{\lambda} + \frac{1}{4} - \frac{q}{\lambda} \frac{x}{\lambda} - \left( \frac{x}{\lambda} \right)^2 \right], \quad (5)$$

where the coefficients  $p$  and  $q$  are written as

$$p = 2E \frac{(1+A)/4 + P + E/\lambda}{A + (1+A)(P + E/\lambda)} \quad (6)$$

and

$$q = E \frac{1-A}{A + (1+A)(P + E/\lambda)}. \quad (7)$$

The parameters  $A=K_-/K_+$  and  $P=K_p/K_+$  characterize the asymmetry of the attachment-detachment kinetics and the step permeability, respectively, both being taken relative to the right-hand step coefficient. The significance of the inverse of  $K_+$ ,

$$E = \frac{1}{K_+} = e^{E_+/kT} - 1, \quad (8)$$

is its equivalence to the kinetic length, which characterizes the rate-limiting step continuously between the DL ( $E=0$ ) and ADL ( $E=\infty$ ) extreme limits. In the classical case of complete impermeability ( $P=0$ ) and symmetric attachment-detachment kinetics ( $A=1$ ),<sup>1</sup> Eqs. (6) and (7) yield  $p=E$  and  $q=0$ . In the diffusion limit,  $E=0$ , one obtains the familiar equation valid for complete condensation and fast attachment-detachment kinetics,<sup>12</sup>

$$n = n_{\text{eq}} + \frac{R}{2D_s} \left( \frac{\lambda^2}{4} - x^2 \right), \quad (9)$$

which is symmetric about a peak at the middle of the terrace.

The first nuclei at a given temperature and a terrace width are formed around the position of the maximum of the adatom concentration.<sup>5</sup> From Eq. (5), the position and the value of the latter are given by

$$x_{\text{max}} = -\frac{q}{2} \quad (10)$$

and

$$n_{\text{max}} = n_{\text{eq}} + \frac{R\lambda^2}{2D_s} \left[ \frac{p}{\lambda} + \frac{1}{4} \left( 1 + \frac{q^2}{\lambda^2} \right) \right]. \quad (11)$$

Note that the position of the maximum depends on the temperature. At given values of the step-edge barriers  $E_+$ ,  $E_-$ ,

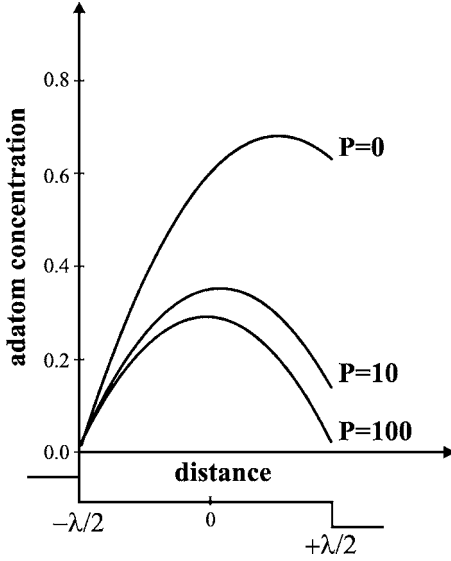


FIG. 2. Influence of the step permeability on the profile of the adatom concentration in units of  $R\lambda^2/2D_s$  ( $E_+ = 0.5$  eV,  $E_- = 0.2$  eV,  $T = 800$  K).

and  $E_p$ , the adatom concentration profile is symmetric at sufficiently high temperature and becomes more and more asymmetric as the temperature decreases.

The asymmetry parameter  $A$  can be either greater or smaller than unity. The case  $A > 1$  means inhibited incorporation of atoms to the right-hand descending step or a normal Ehrlich-Schwoebel (ES) effect. The opposite case ( $A < 1$ ) means an inhibited incorporation of atoms into the left-hand ascending step or an inverse Ehrlich-Schwoebel effect. It can be shown that increasing  $A$  above unity (or decreasing  $A$  below unity) leads to a shift of the maximum of the adatom concentration to the right-hand (left-hand) step edge when  $E > 0$ . The concentration may even possess maxima at the step edges in the ADL regime as the limits  $A \rightarrow 0$  ( $p = \lambda/2$  and  $q = \lambda$ ) and  $A \rightarrow \infty$  ( $p = \lambda/2$  and  $q = -\lambda$ ) are approached. The influence of  $A$ ,  $E$ , and  $P$  on the concentration mimics the behavior of the solution to the diffusion equation inclusive of a desorption term which encompasses the case of incomplete condensation.<sup>5</sup>

Increasing  $P$  shifts the maximum backward to the middle of the terrace, making the population of adatoms symmetric (Fig. 2). The reason is that  $q \rightarrow 0$  and, in turn,  $x_{\max} \rightarrow 0$ . Then, in the limit  $P = \infty$ ,  $n_+ = n_- = \text{const} = n_{\text{eq}} + (R\lambda/D_s) [E/(1+A)]$ . The latter can be easily verified by applying  $x = \pm\lambda/2$  to Eq. (5) and making use of Eqs. (6) and (7).

In the limiting cases  $A = 0$  ( $E_+ = 0$ ) and  $A = \infty$  ( $E_- = 0$ ), the maxima are located at the step edges  $x_{\max} = \pm\lambda/2$ , the expression in the square brackets in (11) is equal to unity, and the maximum adatom concentration achieves its highest value  $n_{\max} = n_{\text{eq}} + R\lambda^2/2D_s$ .

The rate of nucleation reads<sup>6,12</sup>

$$J = KD_s n^{i^*+1}, \quad (12)$$

where  $K$  is given by

$$K = \alpha^* N_0^{-(i^*-1)} \exp\left(\frac{E_{i^*}}{kT}\right),$$

and where  $i^*$  is the critical nucleus size,  $\alpha^*$  is the number of ways an atom can join the critical nucleus,  $E_{i^*}$  is the energy gain when a critical nucleus is formed from single atoms, and  $N_0 = 1/a^2$  is the atomic density of the surface.

Substituting  $n_{\max}$  into Eq. (12) (neglecting  $n_{\text{eq}}$ ) gives the nucleation rate at the position of maximum adatom concentration,

$$J_{\max} = KD_s \left(\frac{R\lambda^2}{2D_s}\right)^{i^*+1} \left[\frac{p}{\lambda} + \frac{1}{4}\left(1 + \frac{q^2}{\lambda^2}\right)\right]^{i^*+1}. \quad (13)$$

It should be noted that the activation energy for surface diffusion is constant only in the case of homoepitaxy when the surface is assumed unstrained. It was found in the case of heteroepitaxial growth of strained surfaces that the diffusion barrier increases when the strain increases from tensile to compressed values.<sup>18,19</sup> As a result, the nucleation rate will depend also on the strain and nucleation will be preferred near the descending step edges rather than in the middle of the terraces.<sup>11</sup> In the present paper, we consider the homoepitaxial growth and all the results given below are valid for this case.

Integrating  $J_{\max}$  from 0 to the time  $t_1 = N_0/R = 1/F$  required to deposit a complete monolayer gives the density of nuclei which appear around the maximum of the adatom concentration. When studying the onset of nucleation at the critical terrace width, this density is, in fact, equal to one nucleus per area of  $\lambda^2$  ( $J_{\max}/F = 1/\lambda^2$ ). From this, we obtain

$$\lambda^{2(i^*+2)} \left[\frac{p}{\lambda} + \frac{1}{4}\left(1 + \frac{q^2}{\lambda^2}\right)\right]^{i^*+1} = \left(\frac{D}{F}\right)^{i^*} \exp\left(-\frac{E_{i^*}}{kT}\right), \quad (14)$$

where  $D = D_s/a^2$  is the hopping frequency. This is the general equation for the critical terrace width for step flow growth which takes into account both the attachment-detachment asymmetry and the step permeability under condition of a complete condensation. It can be easily analyzed to reveal the limiting cases.

It is immediately seen that when the barrier on either side of the terrace is equal to zero ( $A = 0$  or  $A = \infty$ ), the expression in the square brackets in Eq. (14) is equal to unity and the latter reduces to

$$\lambda^2 = \left(\frac{\nu}{F}\right)^{\chi_D} \exp\left(-\frac{E_{i^*} + i^* E_{\text{sd}}}{(i^* + 2)kT}\right), \quad (15)$$

where

$$\chi_D = \frac{i^*}{i^* + 2} \quad (16)$$

is the scaling exponent characteristic for diffusion-limited growth.<sup>6</sup> We conclude that when at least one of the barriers is equal to zero, the kinetic regime is ruled out. Diffusion-limited regime is only allowed as the adatoms join the step with zero barrier upon sticking.

With decreasing coefficient of step permeability (Fig. 3),

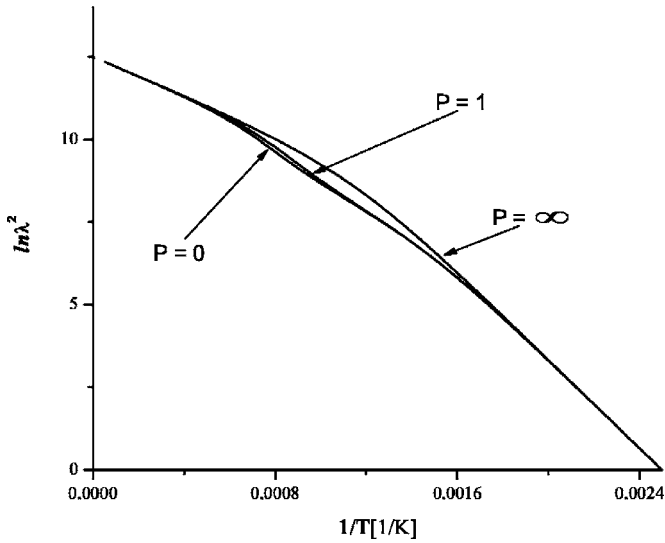


FIG. 3. Effect of the step permeability on the shape of the Arrhenius plot of  $\lambda^2$ . The values of the coefficient  $P$  are denoted by figures at each curve. In the case  $P=1$ ,  $E_p=E_+$ . When  $P=\infty$ ,  $E_p \ll E_+$ . ( $E_+=0.5$  eV,  $E_-=0.2$  eV,  $E_{sd}=0.75$  eV,  $i^*=1$ .)

the curves  $\ln \lambda^2$  vs  $1/T$  show a downward deviation at intermediate temperatures. This behavior continues as long as the coefficient  $P$  is comparable with the remaining parameters and the adatom concentration profile is asymmetric. Once it becomes much larger than the others ( $P \rightarrow \infty$ , completely permeable steps), the adatom concentration becomes symmetric ( $q=0$ ) and the straight line with a slope valid for DL growth smoothly switches over to the other extreme of ADL growth at low temperature. What is more important is that the step permeability does not affect both limiting cases at sufficiently high and low temperatures. Thus, when comparing the theory with experimental data we can neglect the effect of the step permeability when the observations are performed at sufficiently high or low temperatures.

At sufficiently high temperatures,  $p$  and  $q^2$  are small compared with  $1/4$  and unity, respectively, and Eq. (14) turns into

$$\lambda^2 = 4^{(i^*+1)/(i^*+2)} \left(\frac{\nu}{F}\right)^{\chi_D} \exp\left(-\frac{E_{i^*} + i^* E_{sd}}{(i^*+2)kT}\right), \quad (17)$$

which is valid for diffusion-limited regime.

Decreasing the temperature leads to a sharp increase of the values of  $p$  and  $q^2$ . At sufficiently low temperatures,  $p \gg q^2$  owing to the existence of the parameter  $E$  in the numerator in  $p$  and  $(1+q^2/\lambda^2)/4$  can be neglected with respect to  $p/\lambda$ . Then, Eq. (14) turns into the equation valid for the kinetic regime,

$$\lambda^2 = \left(\frac{\nu}{F}\right)^{\chi_K} \exp\left(-\frac{2[E_{i^*} + i^* E_{sd} + (i^*+1)E_{\pm}]}{(i^*+3)kT}\right), \quad (18)$$

where

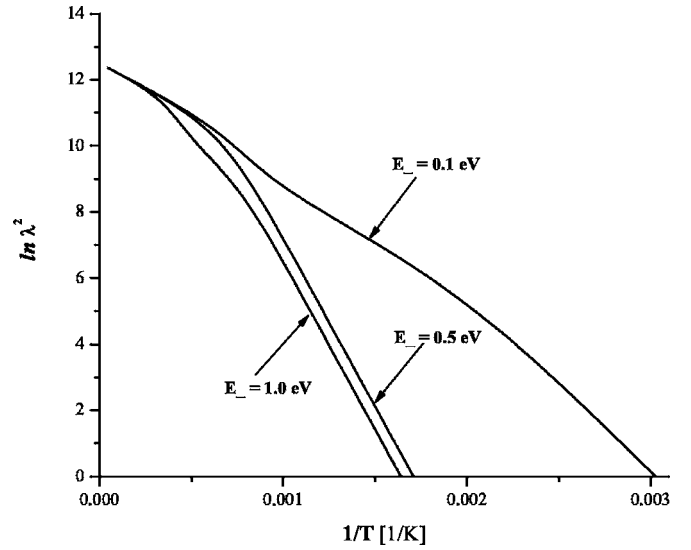


FIG. 4. Arrhenius plot of  $\lambda^2$  at constant right-hand barrier  $E_+=0.5$  eV and different left-hand barriers  $E_-$ . The values of the latter are shown by figures in units of eV at each curve. Note the absence of intermediate kinetic regime at the symmetric attachment-detachment kinetics ( $A=1$ , the curve denoted by 0.5) and the equal slopes of the ADL asymptotes with the values of  $E_-=0.5$  and 1.0 eV. The latter is due to the fact discussed in the text that always the smaller barrier enters the exponential of Eq. (18).  $P=0$  for all curves.

$$\chi_K = \frac{2i^*}{i^*+3} \quad (19)$$

is the scaling exponent which is characteristic for the attachment-detachment limited growth.<sup>20</sup> The slope now includes one of the step-edge (ES) barriers  $E_+$  or  $E_-$ . The barrier  $E_-$  is involved when  $A > 1$  (under conditions of normal ES barrier) and vice versa,  $E_+$  is involved in the case of inverse ES barrier ( $A < 1$ ). This means that the lower barrier is always involved. The reason for the above is that in the case of  $A > 1$  ( $E_+ > E_-$ ), both  $E$  and  $A$  increase with decreasing temperature and  $p \rightarrow E/A = \exp(E_-/kT)$ . In the opposite case ( $E_+ < E_-$ ),  $E$  increases and  $A$  decreases with decreasing temperature. Then,  $p \rightarrow E = \exp(E_+/kT)$ . This behavior is shown in Fig. 4.

Figure 4 demonstrates in addition the effect discussed above. The transition from diffusion limited to attachment-detachment limited regime with decreasing temperature is direct only in the case of symmetric attachment-detachment kinetics ( $A=1$  and  $q=0$ ). The curve consists only of the two asymptotes for the DL and ADL kinetics. If  $A \neq 1$ , there is an additional kinetic limited regime at intermediate temperatures. The latter is due to the fact that in this temperature interval,  $q^2$  is comparable to  $p$  and takes part in determining the critical terrace width. When  $A=1$ ,  $q=0$  and the intermediate kinetic regime disappears.

We examine further the experimental data about the critical terrace width on Si(111)( $7 \times 7$ ) (Ref. 5) in the light of the above considerations. It has been found that  $A=1.8$  cm<sup>2</sup> and



$E=2.0$  eV at  $T>750$  K, and  $A=4.7\times 10^{-8}$  cm<sup>2</sup> and  $E=0.9$  eV at  $T<750$  K.

We treat first the high-temperature data. The values  $E_{sd}=0.75$  eV,<sup>21</sup>  $F=0.05$  ML/min,<sup>5</sup>  $\nu=1\times 10^{12}$  s<sup>-1</sup>, and  $a=3.84\times 10^{-8}$  cm have been used. We find  $\chi=1.005$ . This means a clear kinetic regime with  $i^*=3$ . Obviously, the diffusion regime should be excluded as this means infinitely large nucleus. In the lower-temperature case, one obtains  $\chi=0.498$ . The value of 0.5 means either a diffusion regime with  $i^*=2$  or a kinetic regime with  $i^*=1$ . The diffusion regime is ruled out for a simple reason. If a kinetic regime is observed at higher temperatures, one cannot expect a diffusion regime at lower temperatures.

We conclude that the low-temperature nucleation takes place in a kinetic regime with  $i^*=1$ . Then, from  $0.5(E_{sd}+2E_{\pm})=0.9$  ( $E_1=0$ ) we find the value of  $E_{\pm}=0.525$  eV. As mentioned above, this should be the value of the lower barrier  $E_+$  at the descending step. Chung and Altman established that  $P=0$  and  $x_{max}/\lambda=-0.022$ .<sup>2</sup> In other words,  $A<1$  and an inverse Ehrlich-Schwoebel effect should exist. Then, by using Eqs. (10) and (7) we find  $A=0.87$  and  $E_-=0.535$  eV. We make use of this estimate in order to evaluate the binding energy  $E_3$  from the slope of the high-temperature branch. From  $(1/3)(E_3+3E_{sd}+4E_+)=2.0$  eV, we find  $E_3=1.65$  eV. The latter seems to be out of agreement with bond counting arguments which require a value proportional to the bond energy of Si which is of the order of 2.0 eV.<sup>22</sup> Unfortunately, we cannot say anything about the structure of the nucleus and the place of its formation on the  $7\times 7$  unit cell.

In conclusion, we derived a general expression for the critical terrace width for step flow growth accounting simultaneously for step permeability and the asymmetric incorporation of atoms to ascending and descending steps. It covers both cases of kinetic and diffusion regimes of growth at low and high temperatures. It was found that when at least one of the excess step-edge barriers is equal to zero, only diffusion-limited regime is allowed. The step permeability does not influence both limiting cases at sufficiently high and low temperatures but affects the Arrhenius plot between them. By comparison with experimental data we conclude that the nucleation process at Si(111) $7\times 7$  surface in the interval 700–850 K takes place in a kinetic (attachment-detachment limited) regime. The nucleus consists of one atom below 750 K and of three atoms above this temperature. The values of 0.525 and 0.535 eV (inverse Ehrlich-Schwoebel effect) have been estimated for the energy barriers for incorporation of adatoms into descending and ascending steps, respectively. Making use of these estimates, the binding energy of three-atom cluster has been evaluated from the high-temperature data to be 1.65 eV.

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