Cluster-size distribution during epitaxial growth from the vapor on strongly misoriented surfaces

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Crystal growth in molecular-beam-epitaxy conditions on misoriented surfaces is described with a nonlinear model. Adsorbed atoms may annihilate through collisions with adatoms and existing clusters. Clusters grow only by attachment of single atoms. The conditions under which step flow dominates are displayed as a function of the growth parameters. The cluster density is of the same order or larger than the adatom density. Medium-sized clusters of about 10 atoms will be important also in the conditions most favorable for two-dimensional growth.

I. INTRODUCTION

The microscopic kinetics of epitaxial growth from the vapor in molecular-beam epitaxy (MBE) has received renewed interest recently.[1-3] Because of the very large supersaturations, the classical Burton, Cabrera, and Frank (BCF) (Ref. 4) theory has been modified to take care of step movement.[5,6] More recently, lateral interactions have been included.[1,2,7] This leads to a nonlinear diffusion equation, when two-atom cluster building is considered. This equation is coupled with additional nonlinear equations, if higher-order nuclei are also included.[3] These works emphasize the calculation of the adatom density along the terraces as well as the total cluster density along them.

This paper provides the nonlinear diffusion equation for adatoms coupled with the rate equations for all nuclei in the steady state. They are solved to provide the cluster-size distribution as a function of the growth parameters.

Section II describes the growth model and constructs the full set of nonlinear equations. In Sec. III the real growth conditions are used to approximate the equations. The magnitudes of physical interest for evaluating the deviations from perfect two-dimensional epitaxy are discussed in Sec. IV. In Sec. V, numerical solutions are provided, characterizing the growth modes in the relevant parameter space. A criterion for estimating an epitaxial temperature is also derived. Sections VI and VII provide a discussion and comparison with previous work, as well as the conclusions.

II. GROWTH MODEL

A. Diffusion model

Let us consider a misoriented surface, h being the distance between steps. They are assumed to be monatomic and periodic. An atomic flux F (units of $m^{-2}\ s^{-1}$) impinges onto the surface with a sticking coefficient of unity. Adsorbed atoms (adatoms) migrate with a surface diffusion coefficient D that follows an Arrhenius behavior:

$$D=n_0^{-1}\nu e^{-E_d/kT}, \tag{1}$$

where n_0 is the surface density of sites about $10^{19}\ m^{-2}$ [$6.7\times10^{18}\ m^{-2}$ for the silicon (100) surface]. E_d is the activation energy for a jump between neighboring sites (about 1 eV) and ν is a vibrational frequency near $10^{13}\ s^{-1}$.

According to Ref. 4, let us define E_v as the adatom desorption energy from the surface into the vapor. Then it follows that the mean lifetime τ of an atom on the surface is

$$\tau=\nu^{-1}e^{E_v/kT}. \tag{2}$$

Additionally, its mean displacement or diffusion range x_0 is given by $x_0^2=D\tau$.

B. Adatom conservation

In this model, adatoms diffuse until they reevaporate or collide. Collisions occur with another adatom on the surface or an atom coming from the beam, as well as with existing clusters or the step edges. It is supposed that the Burton, Cabrera, and Frank (BCF) growth mode is only slightly perturbed and collisions with steps dominate.

Under large supersaturation conditions, very far from equilibrium, as it is indeed found in MBE experiments, all clusters are stable (supercritical[8]) and will not detach adatoms. Therefore cluster decay is not considered.

Let $n_i(x)$ (units of m^{-2}) be the surface density of i-sized nuclei ($n_1=n$), where x is the coordinate measured along the terrace between two steps. Actually, these densities should depend on both surface coordinates, that is, $n_i=n_i(x,x')$ (see Fig. 1). Because of the symmetry of the parallel train of steps, however, the dependence on the second axis x' (along the edges), is averaged as follows:

$$n_i(x)=\frac{1}{L}\int_0^L n_i(x,x')dx', \tag{3}$$

where L is a distance small enough on a macroscopic scale, but large enough to contain many clusters. The numerical calculations will show that this distance amounts to several thousands of atomic positions.

Let σ_i be a *capture number,* which represents the efficiency of an i cluster to capture adatoms from the surface. σ_i is roughly proportional to the cluster perimeter

and should increase with $i^{1/2}$ for two-dimensional islands and with $i^{1/3}$ for the three-dimensional situation. For the square lattice, σ_1 should be near 4.

Let σ_i' be the cluster efficiency for trapping atoms from the beam. It is proportional to the cluster area at its bottom, and should increase with i and $i^{2/3}$ for the two- and three-dimensional situations, respectively. For the square lattice, σ_1' is about 5.

In steady-state conditions the steps move to the right with velocity v (see Fig. 1), which is to be calculated. The adatom density n vanishes at both step edges,[4] and the cluster density n_i, $i \geq 2$, vanishes at the recently formed advancing step edge, because clusters have had no time to grow up. Under these conditions, in a reference system moving with the step velocity $\mathbf{v}$, there is a time-independent current $\mathbf{J} = -D\nabla n - n\mathbf{v}$, whose divergence equals the net adatom creation rate.

Adatoms are created because of the impinging flux F, which is corrected to take into account the fraction impinging onto clusters or adatoms. They may also annihilate because of other contributions leading to the following rates: (i) $n\tau^{-1}$ (local) desorption rate from the surface into the vapor; (ii) $2nFn_0^{-1}\sigma_1'$ represents the loss of *both* an adatom and an impinging atom because of a collision; (iii) $2D\sigma_1 n^2$ loss of two adatoms because they collide to form a cluster; (iv) $Dn\sum_{i=2}^{\infty}\sigma_i n_i$ loss of adatoms due to collisions with already existing clusters; and (v) $Fn_0^{-1}\sum_{i=2}^{\infty}\sigma_i' n_i$ fraction of the impinging beam, which does not generate adatoms because they fall directly onto clusters. Therefore, the conservation equation for the adatom density reduces to the one-dimensional form

$$-D\frac{d^2n}{dx^2} - v\frac{dn}{dx} = F - \frac{n}{\tau} - \frac{2F\sigma_1'}{n_0}n - 2D\sigma_1 n^2 - Dn\sum_{i=2}^{\infty}\sigma_i n_i - \frac{F}{n_0}\sum_{i=2}^{\infty}\sigma_i' n_i \ . \tag{4}$$

C. Cluster conservation

All polyatomic nuclei are assumed to be nonmobile. Therefore, there is no cluster diffusion current in the moving system, but only a drift current $\mathbf{J}_i = n_i\mathbf{v}$. Creation and annihilation of clusters take place by attachment of single adatoms from the surface or atoms from the beam (although polyatomic clusters may already be present in the beam itself). The cluster conservation equation for the i clusters with $i \geq 2$ is then

$$-v\frac{dn_i}{dx} = \sigma_{i-1}Dnn_{i-1} - \sigma_i Dnn_i + \frac{F}{n_0}\sigma_{i-1}'n_{i-1} - \frac{F}{n_0}\sigma_i' n_i \ . \tag{5}$$

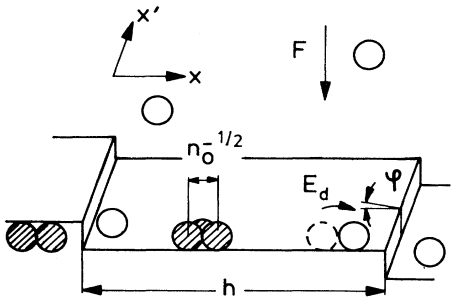

FIG. 1. Advancing monatomic step on a terrace of length h, showing impinging atoms, adsorbed atoms, and clusters, as well as a diatomic cluster incorporated into the crystal.

D. Total cluster density

Finally, an equation for the total cluster density $N(x)$ will be derived. Let us define

$$N(x) = \sum_{i=2}^{\infty} n_i(x) \ . \tag{6}$$

By adding the whole set of Eqs. (5), it follows that

$$-v\frac{dN}{dx} = D\sigma_1 n^2 + \frac{F}{n_0}\sigma_1' n \ . \tag{7}$$

The last result uses the telescopic property assuming that $n_i \to 0$ as $i \to \infty$. Equations (4), (5), and (7) were used in Ref. 7, although no numerical solution was given there. They completely determine the growth problem. The introduction of the total cluster density is based on Zinsmeister's theory of steady-state nucleation.[9] It will be shown that in the experimental conditions found in MBE, it is possible to solve the problem for n and N without solving the equations for n_i.

III. REAL GROWTH CONDITIONS

A. Step velocity

The solution of the coupled equations (4), (5), and (7) is somewhat complicated because the step velocity $\mathbf{v}$, which appears as a parameter, is determined by the solution itself. In most MBE experiments, however,[10] the distance h between steps is much less than the diffusion length x_0. This means that almost all adatoms will meet a step and have no possibility to reevaporate (a situation called *total condensation*). This effect is reinforced if nucleation is included. As a consequence, the term $n\tau^{-1}$ in Eq. (4) can be neglected.

The number of atoms falling onto a terrace in a time t, per unit of length (measured along the steps) is Fht and must equal the number of atoms incorporated to the step in this time, which is the "area" $\mathbf{v}t$ divided by the area assigned to an adsorption site, n_0^{-1}. It follows that in the regime of complete condensation the step velocity is $\mathbf{v} = Fhn_0$ and does not depend on the temperature.

B. Reduced variables

Thinking in the numerical solution it is useful to introduce a nondimensional distance y as $y = x/h$, as well as nondimensional cluster densities z_i by

$$n_i = F\tau\frac{h^2}{x_0^2}z_i \tag{8}$$

and Z by $N=F\tau(h^2/x_0^2)Z$. (The scaling of N used in this work is different from that used in Ref. 3).

In addition, let us define the following nondimensional parameters:

$$b=\sigma_1 n_0 h^2 \tag{9}$$

and

$$a=\frac{F\tau}{n_0^2 x_0^2 \sigma_1}=\frac{F}{n_0 \nu \sigma_1}e^{E_d/kT} . \tag{10}$$

With these definitions, Eqs. (4), (5), and (7), after neglecting the desorption term $n\tau^{-1}$, take the form

$$\frac{d^2z}{dy^2}+ab\frac{dz}{dy}=-1+2ab^2z^2+2ab\sigma_1' z + ab^2 z\sum_{i=2}^{\infty}\frac{\sigma_i}{\sigma_1}z_i+ab\sum_{i=2}^{\infty}\sigma_i' z_i , \tag{11}$$

$$\frac{dz_i}{dy}=bz\left[\frac{\sigma_i}{\sigma_1}z_i-\frac{\sigma_{i-1}}{\sigma_1}z_{i-1}\right]+(\sigma_i' z_i-\sigma_{i-1}' z_{i-1}) , \tag{12}$$

and

$$\frac{dZ}{dy}=-bz^2-\sigma_1' z . \tag{13}$$

Notice that, for $i=1$, the ratio $n_i/n_0=abz_i$ is the adatom surface coverage, i.e., the fraction of the surface sites occupied by adatoms. Additionally, in this work, the surface coverage by i-sized clusters is defined by the ratio n_i/n_0, which always remains several orders of magnitude lower than 1. Because a nucleus occupies several surface sites, this ratio differs from the fraction of the surface actually covered by i-sized nuclei. The latter also depends on the unknown cluster shape.

C. Meaning of the parameters

1. Meaning of b

A short discussion about the parameters a and b is worthwhile. Parameter b depends principally on the misorientation of the surface. Since the distance between adsorption sites is about $n_0^{-1/2}$ and is very near to the height of a monatomic step, b can be expressed in terms of the misorientation angle φ as $b=\sigma_1\cot^2\varphi$ (see Fig. 1). This parameter does not depend on the temperature, except by the possible variations in σ_1. For a given substrate, b is fixed and cannot be changed during the MBE experiment. The lowest value for b can be estimated by accepting that the concept of a misoriented substrate is still valid for, say, a terrace length about five times the atomic height. If a value of 4 for σ_1 is used, it follows that $b=100$.

2. Different parameter elections

The coefficient of the nonlinear term z^2 in Eq. (11) depends on all the growth variables. They are the diffusion energy and temperature as E_d/kT, the impinging flux F, and the misorientation φ. The physical interpretation is quite indirect.

In Ref. 1 this coefficient was written as the product $2\alpha\beta$, with $\beta=b$ and $\alpha=Fh^2/Dn_0$, α depending on the flux, temperature, and misorientation. This has the advantage of providing a physical interpretation for parameter α as the ratio of the time required by adatoms to reach a step edge to the time between arrivals at a surface site. Increasing this parameter increases the surface density and nucleation events.

3. Meaning of a

From a different point of view, it is very convenient that the nondimensional parameters depend on the lowest number of experimental variables as possible. Parameter a, as defined in this paper, depends only on the flux F and temperature T (for a given E_d). Notice that the time $\nu^{-1}\exp(E_d/kT)$ is the mean time between diffusion events and can be regarded as the relaxation time into a lattice position. The time $(F/n_0)^{-1}$ is the mean time between arrivals at a site. Except for the term σ_1, a is the ratio between both times. A small value for a means that adatoms have time to relax into a lattice and to diffuse between arrivals.

A value of a approaching unity means that adatoms are covered before they relax. As it was stated in Ref. 8 and has been further exploited in Refs. 11 and 3, this leads to amorphous growth. (This conclusion does not depend on the surface misorientation.)

D. Capture numbers

At this stage the growth problem can be numerically solved by taking some cutoff value for the cluster size i and selecting two- or three-dimensional island formation, which defines the capture numbers as functions of i. However, if the cluster density decreases with the cluster size, one can concentrate in the smaller nuclei. For them, and beginning with σ_1 about 4, the capture numbers increase rather slowly (sublinearly) with i. It is then sound to approximate all the capture numbers by the same fixed value, that is, $\sigma_i=\sigma_1$ and $\sigma_i'=\sigma_1$. For simplicity, $\sigma_1=\sigma$ will be used. The slight loss of exactitude is greatly compensated for by the generality of the results. With this simplification, all summations in Eq. (11) disappear, leading to

$$\frac{d^2z}{dy^2}+ab\frac{dz}{dy}=-1+2ab^2z^2+2abz+ab^2zZ+ab\sigma Z \tag{14}$$

and

$$\frac{dZ}{dy}=-bz^2-\sigma z . \tag{15}$$

Both equations together with the boundary conditions $z(0)=z(1)=0$ and $Z(1)=0$ can be solved to provide $z(x)$. Then all the cluster densities for $i\geq 2$ can be calculated by iteration.

IV. MAGNITUDES OF PHYSICAL INTEREST

A. Nucleated fraction of the beam

Now let us briefly discuss the physically interesting quantities. The surface current of single adatoms onto a

step, assuming that it has the same capturing efficiency at both edges, is given by the addition of the current $\mathbf{J}=-D\nabla n-n\mathbf{v}$, evaluated at the beginning and at the end of a terrace, where the adatom density vanishes. The fraction γ_1 of the impinging beam that diffuses as single adatoms to the steps in a nondimensional quantity less than unity (it tends to unity for perfect layer-by-layer epitaxy), which is given by

$$\gamma_1=\frac{dz}{dy}_{y=0}-\frac{dz}{dy}_{y=1} . \tag{16}$$

The fraction γ_0 of the beam that is incorporated as preformed nuclei into the steps is then the difference $\gamma_0=1-\gamma_1$. This quantity turns out to be of central importance for two-dimensional epitaxy: if it is small, step flow dominates. If γ_0 approaches unity, nucleation dominates and the formalism has no physical relevance (there is no such thing as step flow). For intermediate values of γ_0 there is competition between step flow and nucleation. The consequences for the overgrowth will depend on the relation between cluster and substrate orientation.

B. Fraction of *i* clusters

Similarly, the current of nuclei of i atoms to a step at the time they are incorporated is $\mathbf{J}_{i(x=0)}=\mathbf{v}n_{i(x=0)}$. The current of *atoms* included in i-sized nuclei is $i\mathbf{J}_{i(x=0)}$. The fraction of the impinging beam that is incorporated in i-sized nuclei is easily shown to be $\gamma_i=iabz_{i(y=0)}$.

The total cluster current into a step, γ_∞, is evaluated in a similar way, leading to $\gamma_\infty=abZ(y=0)$. This expression is normalized to the impingements, and therefore is less than unity (and less than γ_0).

C. Average cluster size

The quantities γ_0 and γ_∞ can be calculated without solving for z_i. This allows one to calculate the average cluster size r as the ratio $r=\gamma_0/\gamma_\infty$.

The magnitude r is a measure of the dominant cluster size at the time the nuclei are absorbed by the advancing step. It can be used as a control for the validity of the assumptions regarding constant capture numbers: if for a given set of growth parameters r is large, say 100, the dependence of the capture numbers on the cluster size becomes important.

V. NUMERICAL SOLUTIONS

A. General considerations

1. *Adatom conservation*

As can be seen in Eq. (14), the dominant nonlinearities in the differential equations are z^2, representing adatom interactions, and zZ, representing adatom-cluster interactions. The coupling parameter g is then given by $g=ab^2$.

As was shown in a previous work,[3] g is a small parameter. Parameter b is a large one (larger than 100). Therefore, the coefficient of the derivative dz/dy in Eq. (14) is small and can be neglected in most situations. Physically, it means that the step movement does not modify the adatom density, as was already stated.[6,2]

The same reasoning can be applied to the term $2abz=2(g/b)z$, which is almost always much smaller than ab^2z^2. This means that collisions between adatoms are more frequent than direct impingement from the beam onto adatoms. The same can be said about beam-cluster collisions.

2. *Cluster conservation*

The linear term σz in Eq. (15) is negligible compared with the quadratic one, except very near the ends of the terraces.

All these terms involve no additional difficulty from the point of view of the numerical solutions. Therefore these are carried out for the complete equation (14).

3. *BCF limit*

When all terms involving step movement or nucleation are neglected in Eq. (14), it reduces to $d^2z/dy^2=-1$. This is the problem already solved by Burton, Cabrera, and Frank[4] for the close step approximation, with the solution $z_{\mathrm{BCF}}(y)=\frac{1}{2}y(y-1)$.

The preceding discussion suggests that the adatom annihilation in Eq. (14) is controlled by the parameter g, and the behavior of the clusters is controlled by b.

B. Solving procedure

The solution of Eqs. (14) and (15) was carried out with a Micro VAX 3600 computer using standard IMSL routines (DBVPFD release 10). Fixed parameter values are the capture number $\sigma=4$ and the step height of one atom. The same procedure was used to calculate $z_i(y)$.

The inclination parameter b varies from 10 to 10 000, corresponding to misorientations from 32.3° to 1.14°. The physically meaningful values of b are larger than 100 (misorientation of 11.3°). The upper limit is given by the mean cluster size, which becomes too large if b is larger than 100 000 (0.36°). The values of the nucleation parameter g vary from 0.001 to 5. The results show that the step flow does not exist for g larger than unity.

Plots of the functions γ_0, γ_∞, and r as a function of the g parameter for fixed values of b were already published[3] (where they were called γ_2, γ_1, and r, respectively). Although these calculations were done for a *biatomic* step considering only the dominant nonlinear terms, no important difference is seen: γ_0 and γ_∞ increase monotonously with g, while the cluster size depends almost uniquely on b, increasing very slightly as g *decreases.*

C. Nucleated fraction of the beam

Much more information can be obtained from Fig. 2, where level curves of constant γ_0 have been depicted as a function of the parameters b and g. The quantity γ_0 can be used to predict if the growth process will be two dimensional (step flow) or not (nucleation).

As has been pointed out in other works, there is no

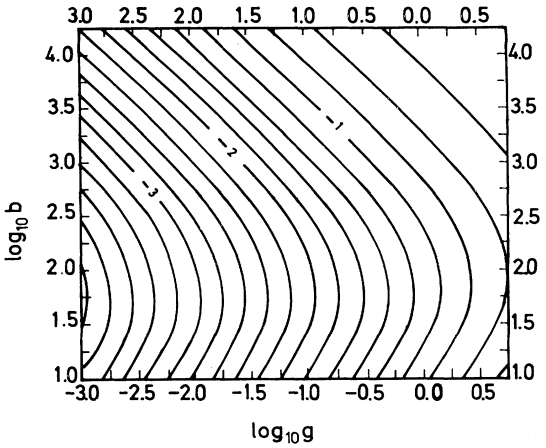

FIG. 2. Fraction of the atomic beam γ_0 that condenses as polyatomic clusters into the steps, as a function of the nondimensional parameters $b=\sigma n_0 h^2=\sigma\cot^2\varphi$ and $g=ab^2=(\sigma F)(n_0\nu\cot^4\varphi)$. $\log_{10}\gamma_0$ is displayed as level curves of constant value. The region at the left side of the level $\log_{10}\gamma_0=-2$ corresponds to the step-flow regime.

sharp transition between layer-by-layer epitaxy and distorted growth.[2,3,12] A rather arbitrary but physically sound criterion was proposed[2] for assuming an epitaxial regime if less than 1% of the beam is condensed into clusters. Similarly, it may be assumed that nucleation dominates if, say, 10% of the beam condenses into clusters. Therefore, the level curves $\gamma_0=0.01$ and 0.1 divide the space parameter (plane g-b) in a step-flow region, a nucleation region, and a transition region, as it is depicted.

From the plot it is inferred that the limiting curve $\gamma_1=0.01$ is quite well described by the relation $bg=ab^3=30$, a very useful approximation for meaningful values of b.

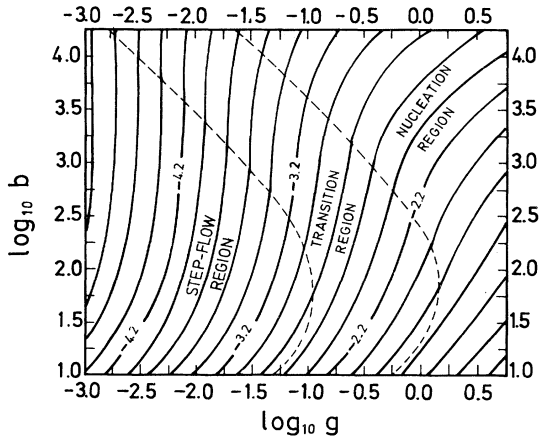

FIG. 3. Total cluster current onto a step edge $\gamma_\infty=abZ(0)$ (normalized to the atom flux onto a terrace). Level curves of constant $\log_{10}\gamma_\infty$ are depicted as a function of the growth parameters $\log_{10}g$ ($g=ab^2$) and $\log_{10}b$. The segmented lines correspond to the levels $\gamma_0=0.01$ and 0.1.

D. Total cluster current

Figure 3 depicts level curves of constant γ_∞, that is, the total number (normalized) of clusters incorporated into the steps, disregarding the number of atoms in them. It is seen that this magnitude depends principally on g and only very slightly on b. This fact was explained through an analytical first-order approximation that gives $\gamma_\infty=0.008ab^2=0.008g$.[7,12]

E. Average cluster size

The last quantity, the average cluster size at the time they meet the step, is depicted as level curves in Fig. 4. In the step-flow region the cluster size is practically independent of the nucleation parameter g: it depends *only* on the misorientation parameter b and *not* on the impinging flux nor the temperature. Hence, b and a describe very different aspects of the growth kinetics.

An important observation is that even for small both g and b the cluster size is larger than 2 and more often about 10. This means that even under favorable growth conditions (i.e., high temperature, low impingement current, and large misorientation), large clusters play an important role. As an example, the calculated average cluster size for $g=10^{-3}$ and $b=500$ ($a=4\times10^{-9}$) is about $r=25$.

The accuracy of the calculation is limited by the large cluster size as b increases: the capture numbers increase with the cluster size and more adatoms are annihilated. Therefore, γ_0 should be somewhat larger than that calculated here. It can also be supposed that the cluster size at large b should be larger, because big clusters capture more adatoms.

Because of the presence of very large clusters, some care is needed for the interpretation of Fig. 4, as well as Fig. 7. In the example above, the distance between steps is nearly 11 atomic spaces for a mean cluster size of 25 atoms. A three-dimensional cluster of this size will have

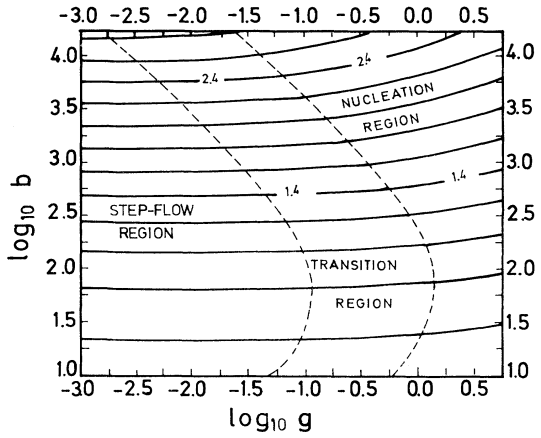

FIG. 4. Average cluster size r at the time the nuclei are captured by the steps. $\log_{10}r$ is depicted as a function of the growth parameters $\log_{10}g$ and $\log_{10}b$. The segmented lines correspond to the levels $\gamma_0=0.01$ and 0.1 ($g=ab^2$).

a base diameter about three atomic positions, and a two-dimensional one about five, occupying a significant portion of the terrace length. Under these conditions the cluster position x turns out to be meaningless. This seems to be an inherent limitation of the continuous model for the description of the discrete nucleation process.

From the physical point of view this only means that the nuclei meet the step edges earlier than assumed in the model. If the cluster and the substrate orientation coincide, this cluster-step collision can be interpreted as a fluctuation of the step shape.

On the other hand, if the cluster orientation is different, a grain boundary may form. It is difficult to predict how it influences the growth at later stages: the defect may be covered and buried by the next layers, or it may propagate upwards, eventually leading to the complete disruption of the layer growth as the overlayer becomes thick enough. Only in the former situation can one speak of steady-state layer growth. In the latter, the quality of the overgrowth decreases with increasing thickness and may turn to be polycrystalline after some critical thickness is reached.

F. Growth temperature

As was mentioned, there is no well-defined epitaxial temperature. By accepting that good epitaxy in many materials is achieved only in the step-flow regime, a quality-dependent growth temperature T_e can be defined in terms of the value of γ_0.

From the plot of Fig. 2 it can be inferred that the level curves of constant γ_0 are very similar to the curves of constant $bg=ab^3$. This was confirmed by plotting the level curves for γ_0 as a function of b and bg, as in Fig. 5. If the b parameter is larger than 100, the γ_0 level curves are roughly similar to the constant bg ones. They are also of nearly constant spacing. Therefore the product $bg=ab^3$ is a natural growth parameter. A numerical fit leads to the roughly approximate expression $\log_{10}\gamma_0 \approx \log_{10}(bg)-3.5$.

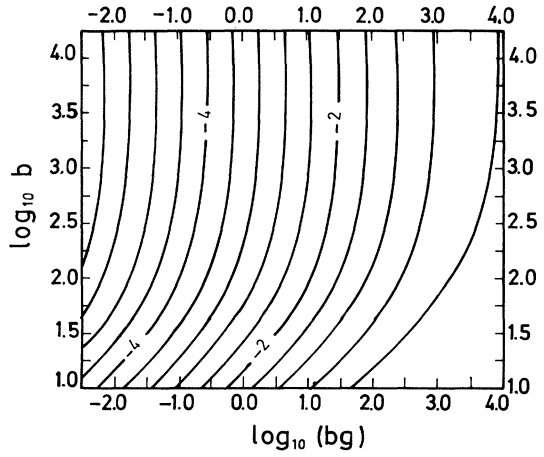

FIG. 5. Fraction of the atomic beam γ_0 that condenses as polyatomic clusters into the steps. Level curves of constant $\log_{10}\gamma_0$ are depicted as a function of the "natural" growth parameters $\log_{10}b$ and $\log_{10}bg$. It is observed that in the physical region $b>100$, γ_0 is nearly a function only of bg, but not of b ($g=ab^2$).

Using the definitions of b, g, and a it follows that

$$T_e \approx \frac{E_d}{k}\left[\log_{10}\left[32\gamma_0(\%)\frac{n_0\nu}{F\sigma^2}\tan^6\varphi\right]\right]^{-1}, \tag{17}$$

where $\gamma_0(\%)$ denotes the percent of the impingement beam incorporated as clusters. Very often the ratio $32/\sigma^2$ approaches unity, providing a simple approximation.

The surface site density is very similar for different materials, nearly 10^{19} m^{-2}. Also the frequency ν is about 10^{13} s^{-1}. The impingement rate changes between different experiments, but because of technical reasons it is often nearer to 10^{19} m^{-2} s^{-1}. Using these illustrative values and assuming that the threshold occurs for $\gamma_0(\%)\approx 1$, one obtains

$$T_e(1\%) \approx \frac{E_d/k}{[32+6\log_{10}(\tan\varphi)]}. \tag{18}$$

G. Approximate expressions

The quality of the overgrowth depends not only on γ_0 but also on the average cluster size r. Therefore, an approximation for r is very useful. From the plot of Fig. 4 a numerical fit gives roughly $\log_{10}r \approx 0.85(\log_{10}b-1)$.

Together with the approximation for γ_0, it follows that

$$\log_{10}\gamma_\infty \approx \log_{10}\gamma_0 + 0.157\log_{10}b - 2.65 .$$

Because of the limited range of b, one can select $b=1000$, leading to $\log_{10}\gamma_\infty \approx \log_{10}g - 2.2$, or $\gamma_\infty \approx 0.006g$. The last expression is roughly similar to the first-order analytical expression.[7,12]

H. Cluster-size distribution

1. *Fraction of the beam in i clusters*

An additional aspect of the growth process is the distribution of the cluster size as a function of i at a fixed position y, in particular at $y=0$, where it is related to the cluster current. Figure 6(a) is a plot of the fraction of the atomic beam condensed as i-sized clusters into the steps, γ_i, as a function of the cluster size i. Different curves correspond to different inclination parameters b, for fixed g. The maximal contribution shifts to larger clusters as the misorientation decreases. The shape of the curves does not depend on g. This is consistent with the independence of the cluster size on g.

2. *Cluster-size distribution*

The current of i-sized clusters, normalized to the impinging current onto the terrace, is displayed in Fig. 6(b), showing a similar behavior. Notice that the maxima are at least in the neighborhood of $i=8$ and increase as b increases.

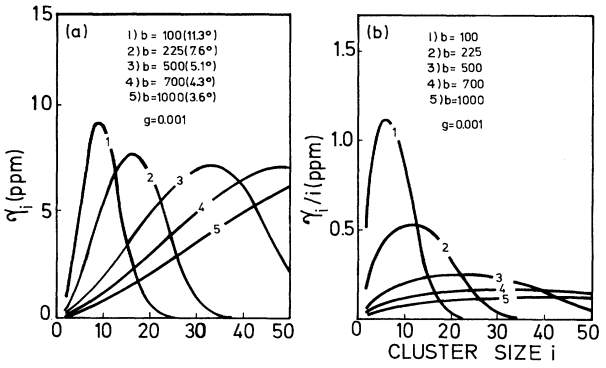

FIG. 6. (a) Plot of γ_i, fraction of the atomic beam that condenses as i-sized clusters into the steps, as a function of the cluster size i. Different curves correspond to different b values. The equivalent misorientation is indicated in parentheses. The curves are plotted for $g=0.001$, but they are nearly independent of g. (b) Plot of the current of i clusters into the steps (normalized), γ_i/i, as a function of i.

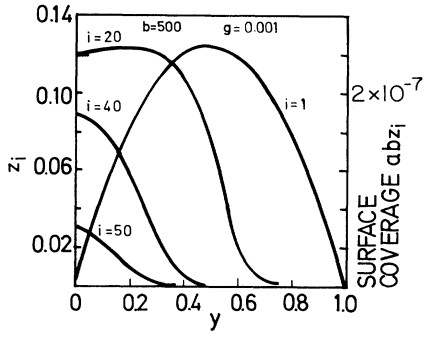

FIG. 7. Reduced surface cluster density z_i as a function of the position on the terrace. The scale on the left is z, the scale on the right corresponds to the real surface coverage n_i/n_0. The growth parameters are $b=500(5.1°)$ and $g=0.001$, a point well inside the step-flow region. The distance between steps amounts to eleven atomic positions.

Physically, this means that the cluster-size distribution *does not depend on the temperature or the impingement rate:* it is only a function of the misorientation.

Even at rather high misorientations ($b=100, \varphi=11.3°$), clusters of size about 8 atoms make the most important contribution. The absolute value of γ_i, however, depends on the temperature and the flux through the g parameter. Curves for different b values scale rather similarly as g changes.

I. Cluster density

Finally, the cluster densities $z_i(y)$ are displayed in Fig. 7 as a function of the position y, for some different cluster sizes. The selected growth parameters are $b=500$ ($\varphi=5.1°$), a realistic misorientation, and $g=0.001$ ($a=4\times10^{-9}$). These values correspond to dominant step-flow conditions.

The value of g affects the scale of the plot, but not its shape. Small clusters, as $i=2$ (not depicted) follow a curve very similar to $z(y)$ (single adatoms), with a maximum near the center of the terrace. Their density decrease as one approaches the step. At about $i=20$ the maximum and the value at the edge are very similar. At $i=30$ the situation is reversed: their is no maximum and the cluster density steadily increases near the advancing edge.

The asymmetry of the curves is explained as follows: One can argue that smaller cluster densities have their maximum value near the maximum of the adatom density, because they are created in this region. The density decreases as one approaches the step edge, because it becomes larger through the capture of adatoms. On the other hand, the density of large clusters increases continuously as one approaches the step, with a correspondingly simple interpretation: they are created as expenses of smaller nuclei and adatoms. The slight asymmetry in $z(y)$ cannot be appreciated in the scale of the figure.

VI. DISCUSSION

A. General

This paper puts in a quantitative form the well-known fact that high temperatures, low growth rates, and a quite large misorientation increase the quality of homoepitaxial layers that require a step-flow mechanism. On the other side, lower temperatures and higher growth rates are desirable for production. Therefore, a compromise must be found, depending on the acceptable level of incorporated nuclei in the overgrowth. For a given substrate (b fixed) and growth conditions (T and F, incorporated in a), this number can be predicted from Fig. 2. This requires an independent knowledge of the diffusion energy E_d, which is only sometimes available and with some uncertainty.

The predicted γ_0 values should be interpreted as a highest limit, because small clusters may rearrange at the time they meet the step and match the bulk structure perfectly. Because of the results depicted in Fig. 6, however, the highest contribution comes from medium or large clusters, which are not likely to rearrange easily.

B. Cluster density

The behavior of the cluster density as depicted in Fig. 7 is, at first glance, somewhat counterintuitive. All cluster densities up to $i=30$ or even larger have maxima similar to the single adatom density. The total reduced cluster density $Z(y)$ was indeed not depicted, because it is more than one order of magnitude out of range in the scale of the figure. This means that *there are many more clusters than adatoms* on the surface, even in conditions at which step flow largely dominates.

Equation (15) provides some help for the understanding of this fact. Notice that the maximum possible value of z is 0.125 and that of σ' is 4. Neglecting the quadratic term, this leads to a maximum value of nearly 0.5 for $Z_{(y=0)}$, effectively larger than $z_{\max}$.

The physical picture can be described as follows: the average distance d between clusters must be larger than $N(0)^{-1/2}$. It is easy to show that $N(0)=n_0\gamma_0 r^{-1}$. In reference to Fig. 7 and using the level curves, the following values are found: $\gamma_0=2.2\times10^{-4}$ and $r=25$. This leads to d larger than 300 atomic positions in any direction, parallel or perpendicular to the steps. This is much larger than the distance between steps, which amounts to 11 atomic positions. Therefore, incoming atoms will meet a step with much higher probability than a nuclei, and step flow dominates despite the large average cluster size.

Incidentally, the mean distance between adatoms for the same growth parameters turns out to be larger than 2000. This is consistent with the large cluster size, because adatom-adatom collisions will be scarce as compared with adatom-nuclei collisions. This can be summarized by stating that adatom-adatom distances are larger than nuclei-nuclei distances, which are larger than the step-step distance. Reciprocally, adatom-step collisions are more frequent than adatom-nuclei collisions (therefore step flow), which are more frequent than adatom-adatom collisions (therefore the large cluster size).

The important feature to remember is that the adatom *current* into the steps is much larger than the cluster current, and that adatom-cluster collisions are rare events as compared to adatom-step edge collisions. The lower adatom density, as compared with the clusters, is only a consequence of their high diffusivity: after arrival, adatoms migrate very quickly to the step edges, while the nuclei remain fixed at their positions.

The cluster density can be higher than the adatom density, but it remains far below the total surface coverage, as depicted in Fig. 7.

C. Effect of the misorientation

The effect of the misorientation, represented by b, and the growth parameters flux F and temperature T, represented by a are very different. In particular, the average cluster size and the cluster-size distribution depend only on b. Large misorientations favor smaller clusters. A consequence is that, for all growth conditions, clusters as large as 10 atoms and much larger must be considered in any calculation.

D. Influence of the large clusters

Early approximations have only considered the adatom collision rate in a linear approximation,[7] or cut the cluster size at two or three atoms.[2] These approximations are useful for calculating the adatom density, but ignore the size distribution. A step-flow regime was predicted[2] if the condition $ab^2=0.01$ is satisfied, by considering the weight of the nonlinear terms in the differential equations.

Differences with the actual calculation can be seen in Fig. 2, where the condition $ab^2=0.01$ corresponds to the vertical line $\log_{10}g=-2$. Roughly speaking, both criteria are similar at very high misorientations, but the latter is not restrictive enough at smaller angles, for which larger clusters are more important. In this situation, the numerical plot of Fig. 2 should be preferred.

E. Limitations of the model

The fact that large clusters are important limits the range of b values for which the model can be directly applied. If b is large (say, misorientation smaller than 1°), the size dependence of the capture numbers must be taken into account. The existence of magic numbers for clusters of especially high stability may also influence the size distribution.

The model does not consider surface reconstruction. Its effect is included in the value of the diffusion energy E_d and, possibly, the surface site density n_0.

VII. CONCLUSIONS

Nucleation on the terraces leads to important deviations with respect to the BCF growth theory. The adatom density is only sightly modified in the step-flow region, but a fraction of the impinging beam condenses as nuclei and not as single adatoms. The size distribution of the clusters depends mainly on the misorientation of the vicinal surfaces, and not on the growth conditions (temperature and growth rate). A criterion for evaluating a quality-dependent growth temperature as a function of the misorientation and flux is provided.

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