Determination of step-adatom kinetics by reflection high-energy electron diffraction during step-flow growth: Theory

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We propose a method for using reflection high-energy electron-diffraction (RHEED) measurements to determine step-adatom kinetics during growth by step flow. Analytical results are obtained for the adatom sticking coefficients at the steps in terms of the steady-state in-phase RHEED specular intensity at two distinct misorientations. Calculated results based on estimated values for these intensities in the step-flow regime indicate the presence of a pronounced barrier at the down step, providing support for previous conjectures. Extension of these results to lower temperatures, including the region below the critical temperature where the majority of RHEED studies reported have been made, should be possible but will involve calculations of a more complicated nature.

I. INTRODUCTION

There has been a great deal of work in recent years[1] concerned with elucidating various aspects of growth on vicinal semiconductor surfaces during molecular-beam epitaxy. The kinetic equation originally introduced by Burton, Cabrera, and Frank[2] (BCF) for growth at low supersaturation and near-equilibrium conditions has been, with appropriate modifications, the basis for much of this work.[3] Here, as is also the case with computer simulations,[4] it has been necessary to make informed guesses regarding the boundary conditions at the steps.[5–7] A variety of choices has been proposed. The most general choice is for unsymmetric conditions at the up- and down-steps.[5,8] More frequently, explicit results are obtained using absorbing boundary conditions. This choice is problematic in the context of a macroscopic description, for which this condition is taken operationally to mean that the adatom density vanishes at the steps.[5–7] Further, when step motion is considered,[1,9,10] this condition is inconsistent since the density profile across the terraces, including the terminating steps, must be anisotropic.[5]

At present there appear to be only a few general rules that can determine the choice of step boundary conditions, and even these are not ironclad. Thus the stability criterion[11] that the up-step collects adatoms more effectively than the down-step, i.e., that there is a barrier to hopping down, has been questioned.[12] This cannot be inferred from growth experiments, since the latter are two dimensional and other criteria must be considered as well.[13] Also, impurity effects in real experiments can alter the criterion as formulated for the modified BCF one-dimensional models.[14,15] It would clearly be very useful to be able to establish a link connecting current experimental capabilities and step boundary conditions, and that is the specific purpose of this paper.

We must emphasize here, as we will do again in Sec. IV, that we cannot verify the procedure we propose at this time by direct comparison with experimental data. What we are able to show is that this leads to reasonable qualitative results, within the context of the BCF description, and that further refinement and extension of these results in conjunction with experimental inquiry are warranted. In Sec. II we briefly discuss the reflection high-energy electron-diffraction (REED) experimental results that are required. We relate these results directly to step boundary conditions in Sec. III, obtaining explicit results in the context of the BCF equation for step-flow conditions; our choice of the latter is due to the relative analytical simplicity of this case, but it would be possible to extend these results to lower temperatures where moving step and islanding effects cannot be neglected.[3,8,10] A discussion of the results obtained, and their limitations, together with our conclusions, follows in Sec. IV.

II. RHEED EXPERIMENTAL INPUT

RHEED is now a routine diagnostic procedure for the *in situ* study of molecular-beam-epitaxy growth kinetics.[16] Despite its ubiquity, there remain questions concerning the comparison of data with theory, and here we choose to directly compare the step surface density, or the adatom density for step-flow conditions, with the in-phase (Bragg) intensity.[1,4,17] Remarkable agreement between experiment and simulation results reported in the recent literature dictate this choice.[4,17] Direct comparison of the results we obtain with real data will not be made, since the theoretical model we choose is for the case of step flow, whereas almost without exception the RHEED data reported in the literature are for temperatures at or below the critical temperature signaling the onset of intensity oscillations due to islanding. A further problem, of less serious nature, faced by the theorist is that RHEED data are generally reported in arbitrary units (a.u.) without a displayed intensity scale, so that access to raw data is necessary.

For a given misorientation, which we represent here as a/L where a^2 is the site density and L the terrace length, the measured in-phase RHEED specular intensity appears to correlate quite well with the step density or, equivalently, at step flow conditions, adatom density across[4,17] the terrace, so that we have

$$I_i(t)=A_i\left|\int_0^1 dx\,[1-2\bar{n}(x,t)]\right|^2 , \tag{1}$$

where $I_i(t)$ is the specular intensity for fixed $\alpha_i \equiv a/L_i$, A_i is also a constant for fixed α_i, and $\bar{n}(x,t)$ is the (dimensionless) adatom density[8] on the terraces. The constant A_i has been considered to be independent of the temperature in previous work,[4,17] but this is not essential for what follows. The important consideration here is that A_i takes into account the complex interplay between surface morphology and the diffraction processes in a systematic way that allows for reproducibility at fixed misorientation and beam flux.

Equation (1) is the basis for what follows. Since $\bar{n}(x,t)$ contains information concerning the boundary conditions, we will be able to relate the latter to the intensity through this relationship, and this is done in Sec. III. As we have shown previously,[3,5-7] it is possible to formulate the boundary conditions explicitly in terms of the sticking coefficients at the steps, and thus Eq. (1) allows us to directly express I_i in terms of these quantities. Since we expect the step kinetics to be asymmetric, i.e., to differ at the up- and down-steps, Eq. (1) then relates the two sticking coefficients to the intensity at a fixed time, but this relationship also involves the unknown A_i.

Two possibilities for simultaneously eliminating the unknown A_i and obtaining two independent equations relating experimentally determined I_i to the two different sticking coefficients suggest themselves. First, using $I_i(0)$, $I_i(\mathrm{ss})$, and $I_i(\mathrm{eq})$, where the last two of these denote the values in the steady state and relaxed, equilibrium state, the first of these can be used to normalize the last two and remove A_i. This offers the advantage of using measurements at a single temperature and misorientation. However, in describing the equilibrium state extreme care regarding the detachment process at the steps is required, since this is no longer overwhelmed by the nonequilibrium processes that dominate in the high supersaturation environment that the source beam creates. To avoid such complications this first approach will not be considered further here. An alternative approach, which we adopt in what follows, is to consider $I_i(0)$ and $I_i(\mathrm{ss})$ at fixed temperature for two distinct misorientations, using the former in each case to normalize the second and thereby eliminate A_i. Since the sticking coefficients most likely will depend on temperature, it is necessary that the temperature remain the same at each misorientation. In Sec. III we determine expressions for the two sticking coefficients in terms of $\bar{I}_i \equiv I_i(\mathrm{ss})/I_i(0)$.

III. STEP STICKING COEFFICIENTS

In previous work[5-7] we have shown that the boundary conditions for the modified BCF equation can be expressed in terms of the step sticking coefficients S_0 and S_L at the left, ascending (up) step at $x=0$ and at the right, descending (down) step at $x=L$, respectively, for a terrace of length L whose coordinate runs from $0 \le x \le L$. For a nondimensionalized adatom density $\bar{n}(x,t) = Dn(x,t)/JL^2$ and $\bar{x}=x/L$, where D is the adatom diffusion coefficient on the terrace and J the source beam strength, the steady-state-modified BCF equation and boundary conditions are[7]

$$n_{xx}+1=0 , \tag{2}$$

$$-h_0 n = n_x, \quad x=0 , \tag{3a}$$

$$h_1 n = n_x, \quad x=1 , \tag{3b}$$

where we now drop all overbars. The values of h_j are given,[5,7] in dimensionless form here, as

$$h_j=(S_j)(2-S_j)^{-1}(2/\pi^{1/2}), \quad j=0,1 . \tag{4}$$

As is usual in describing two- (and three-) dimensional phenomena with a one-dimensional model, we do not expect a precise quantitative description of the actual surface kinetics from Eqs. (2)–(4). However, we do expect that these equations reflect the main qualitative features, and it is in this spirit that we continue and regard our final results.

Solving Eq. (2) together with the boundary conditions, and using this result directly in Eq. (1), we find

$$I_i^{1/2}=1-(h_1+h_0+h_1h_0/\alpha_i)^{-1} \times[2\alpha_i+\tfrac{2}{3}(h_0+h_1)+h_0h_1/6\alpha_i] . \tag{5}$$

For two distinct values of i, Eq. (5) then allows us to determine h_0, h_1 in terms of α_i, I_i, for any distinct set of values of these quantities. The resulting equations for h_0, h_1 are both lengthy and complicated, and need not be written out here since they are implicit in Eq. (5). This equation can be solved, e.g., for h_1, in terms of α_i, $I_i^{1/2}$, and h_0, giving

$$h_1=[2\alpha_i-h_0(1-I_i^{1/2})+2h_0/3] \times[(1-I_i^{1/2})(1+h_0/\alpha_i)-(\tfrac{2}{3}+h_0/6\alpha_i)]^{-1} \equiv F(\alpha_i,I_i,h_0) , \tag{6}$$

and h_0 is then determined by solving

$$F(\alpha_1,I_1,h_0)=F(\alpha_2,I_2,h_0) \tag{7}$$

since the step kinetics should be independent of the misorientation for, e.g., $\frac{1}{20}<\alpha_i<\frac{1}{40}$ so that diffusion is the primary migration mechanism.

IV. CONCLUSIONS

Equations (6) and (7) are the primary results of this paper, and provide the basis for determining step sticking coefficients from RHEED data. As discussed earlier, data relative to the step-flow conditions described by these equations are not available to us, since the primary use of RHEED has been to study the oscillations that occur below the critical temperature, where step flow does not occur. Because of this, we restrict ourselves here to demonstrating the plausibility of the above results by using values of I_i inferred from results in the literature near and at the critical temperature[4,17] together with expected extrapolated changes at increased temperatures and variations in misorientation. Even without the a refinements in the BCF model required to obtain qualita-

tively more precise results [e.g., the numerical factor in Eq. (4) is clearly a model-dependent artifact], this should allow us to answer the important open question of whether there is a barrier at the descending (down) step.

We consider GaAs(001) with misorientations of about 1.6° and 2.3° corresponding to $\alpha_1=\frac{1}{35}$ and $\alpha_2=\frac{1}{25}$; it then follows that $I_1>I_2$, and we take $I_1^{1/2}=0.70$ and $I_2^{1/2}=0.66$ as reasonable estimates for the step flow[4,17] at some undetermined temperature above the critical value in the step-flow regime. For these values we solve Eq. (7) to determine h_0, and then h_1 follows from Eq. (6). We find $h_0=0.80$ and $h_1=0.10$, and then from Eq. (4), $S_0=0.83$ and $S_1=0.09$. As we have emphasized above, the quantitative aspect of these results is only approximate given their model dependence and the values of I_i used to determine them. Still, the qualitative aspect remains creditable, and very clearly indicates the existence of a barrier at the down-step. This is a very important conclusion that agrees with recent results based on an *a priori* assumption of a barrier.

The results obtained here can serve two purposes. First, they provide the motivation for consideration of the more analytically formidable case, below the critical temperature, where islanding occurs[3,8,10] and terrace morphology plays a more significant role in determining the RHEED signal than at step-flow conditions when only adatoms are present. In the latter case, as we mentioned earlier, results obtained with the one-dimensional model used here and in many other studies[1-3,8-11,14] cannot be given a strict literal interpretation. In this regard, the sticking coefficients should be considered as a coarse-grained[8] average across the steps which on a true substrate, as encountered by a RHEED beam, are of a two-dimensional character. The sticking coefficients determined above are therefore not true sticking coefficients, which will vary across the step due to surface tension effects associated with local curvature.[13] In this case then, the RHEED experiment serves the purpose of providing a basis for determining the boundary conditions to use with the one-dimensional model.

In conclusion, we show how the above interpretation of our results can be extended to the probabilistic model[1] that has been used to study terrace size distributions both analytically and by simulation. In this model the migration on the terraces is not explicitly described, and the terrace size evolution is determined solely by the probability p that a deposited adatom is incorporated at the up-step (or at the down-step with probability $[1-p]$). The probability p in the steady state is given by the ratio of the flux at the up-step to the deposited flux, so that from Eqs. (2)–(4)

$$p=h_0(1+\tfrac{1}{2}h_1\alpha^{-1})(h_0+h_1+h_0h_1\alpha^{-1})^{-1}\ . \tag{8}$$

For the cases considered here, $\alpha_1=\frac{1}{35}$ and $\alpha_2=\frac{1}{25}$, we find $p=0.65$, and 0.62, respectively. The dependence of p on α is explicit in Eq. (8), and is a consequence of the enhancement of recapture with increased terrace length.

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