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Step-motion-imposed asymmetry during molecular-beam epitaxy on vicinal surfaces

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Epitaxial crystal growth by molecular-beam epitaxy is frequently described using a modified Burton, Cabrera, and Frank model. Step motion is included in this model through the addition of a convection term and results in an asymmetric density profile across the terraces. The use of macroscopic absorbing boundary conditions at the steps represents an overconstraint since both the kinetics and adatom densities are then equal on both sides of the steps. We make use of a microscopic boundary condition that allows us to properly account for the asymmetry when the steps are absorbing. In the case where the adatom densities at both sides of the steps are equal we determine the relationship that then constrains the step kinetics and obtain specific results for the case where the upstream step is absorbing and the Peclet number is less than 1.

I. INTRODUCTION

The purpose of this paper is to consider some general features of the steady-state adatom density as described by the crystal-growth model of Burton, Cabrera, and Frank[1] (BCF) with modifications appropriate to the conditions of molecular-beam epitaxy (MBE).[2–5] We restrict our discussion to the case where nucleation on the terraces can be neglected,[6] which allows exact analytical results to be obtained. The motivation for this study originated from ongoing work[5] concerned with the case where nucleation effects occur and for which it would be useful to have a clear understanding of the effects resulting from step flow alone. The modified BCF equation that describes this situation includes a convective step flow term, omits the desorption term, and requires the use of nonequilibrium boundary conditions.[2–5] A related equation that includes a desorption term has also been considered.[7] These previous studies have mainly[2–4,7] focused on numerical analysis and not been concerned with the analytical properties of the solution. Except for our own work,[5] the description used has been at the macroscopic level, which limits the amount of detail that can be included in modeling the boundary conditions.

The particular feature that we are interested in is the nature of the asymmetry in the adatom density profile across the terrace that results from the step motion alone or together with asymmetric conditions (kinetics) at the steps. It is clear that as a result of the step motion the profile will be skewed toward the upstream step. As a consequence, if the step kinetics are symmetric the adatom densities at both sides of the steps should differ. Then the use of absorbing boundary conditions[2,4,8] along with a macroscopic description overconstrains the problem, since in that case the kinetics and the step densities (identically zero) are each equal. Our primary purpose here is to resolve this dilemma. We will also consider some related features that result from the step-motion-imposed asymmetry, the location of the maximum adatom density on the terrace, and the relationship describing the constraint on the step kinetics when the adatom densities at the steps are equal. Although we are concerned with qualitative issues here, we also assess the quantitative consequences in order to place our result in a proper perspective.

In Sec. II we formulate the problem and make use of a microscopic description[5,9] that allows us to accurately model the step boundary conditions in terms of the step sticking coefficient; the specific results for the boundary conditions are relegated to the Appendix, since this is a straightforward generalization of work discussed elsewhere.[10] In Sec. III we show that these boundary conditions lead to the proper asymmetry, i.e., unequal adatom step densities. Some numerical results are also given to allow comparison with the results obtained using the macroscopic boundary condition. In Sec. IV, we briefly consider what the implications are for the step kinetics when the adatom densities at the steps are equal, and explicit results are obtained for the case where the upstream step satisfies a microscopic absorbing boundary condition.

II. MODIFIED BCF EQUATION AND BOUNDARY CONDITIONS

The adatom density on the terraces satisfies a modified BCF equation that incorporates step-motion effects through a transformation of the space variable X to a coordinate system moving with the step velocity V. If L is the terrace length, $N(X)$ the adatom density, and F the beam flux, then the steady-state equation in nondimensional form is[2–4]

$$n_{xx}=wn_{x}+1=0, \quad 0\leq x\leq 1 , \tag{1}$$

where $w=FL^{2}(Dn_{0})^{-1}$ is dimensionless, D is the adatom diffusion coefficient, and $n_{0}=a^{-2}$ the terrace site density. The coordinate transformation is $X=(Lx+Vt)$, and the density has been nondimensionalized according to $N=wn_{0}n$. Since $V=FL/n_{0}$, the parameter $w=VL/D$ and can be interpreted as a Peclet number; we will be interested in values of w in the range $0\leq w\leq 1$.

In the microscopic description, Eq. (1) is the lowest-order approximation in a systematic procedure that allows a progressively more detailed description.[9] The lowest approximation includes Fick's law $J=-DN_X$ as a derived relationship, i.e., it is not a separate assumption as in the macroscopic description. Although the equation of transport is the same for both the microscopic and macroscopic descriptions at this level of approximation, there still remains a basic difference as regards the boundary equations that complement this equation. Macroscopic boundary conditions are expressed in terms of n, e.g., for absorption at the steps the adatom density is taken to vanish, $n(0)=n(1)=0$. Microscopic boundary conditions can be expressed in terms of adatoms entering, n_{in}, and leaving, n_{out}, the steps, so that at each step a sticking coefficient $S_i=1-[n_{\text{out}}(i)/n_{\text{in}}(i)]$ can be defined that more accurately characterizes the step kinetics than is possible with a macroscopic description. For absorption there are no adatoms leaving the steps, and $S_i=1$. In the Appendix we show that the general microscopic boundary conditions complementing Eq. (1) are

$$n_x=h_0(L/a)n, \quad x=0 \tag{2a}$$

$$n_x=-h_1(L/a)n, \quad x=1 \ , \tag{2b}$$

where the h_i are functions of the S_i and are given by Eqs. (A4b) and (A5b). The general solution of Eq. (1) is

$$n(x)=B-(A/w)e^{-wx}-(x/w) \ , \tag{3}$$

where the constants A and B follow from Eqs. (2):

$$A=[h_1+h_0+h_0h_1]/[h_0h_1(1-e^{-w})+w(h_1+h_0e^{-w})] \ , \tag{4a}$$

$$B=[w(1+h_1)+(h_0+h_1)+(h_1-w)e^{-w}]/w[h_0h_1(1-e^{-w})+w(h_1+h_0e^{-w})] \ . \tag{4b}$$

Equations (3) and (4) are general; in the next section we consider the case of absorbing steps, $S_0=S_1=1$.

III. ABSORBING STEPS

In the macroscopic description the boundary conditions for absorbing steps are $n(0)=n(1)=0$, and together with Eq. (3) give

$$n_M(x)=(1-e^{-wx})/w(1-e^{-w})-x/w \ . \tag{5}$$

As we discussed in Sec. I, the combination of symmetric step kinetics and equal step densities is in conflict with the expectation that the moving step will not only skew the adatom density profile across the terrace, as described by Eq. (5), but also result in a density discontinuity across the step which Eq. (5) will not predict. Only when the step kinetics are unsymmetric would we expect that the density across the step might be continuous. The microscopic boundary conditions Eqs. (4) provide the basis for resolving this apparent conflict.

When both steps absorb $S_0=S_1=1$,

$$A=(2+L')/[L'+w-(L'-w)e^{-w}] \ , \tag{6a}$$

$$B=[w(1-e^{-w})+L'(1+w)+L'(L'+e^{-w})]/w[L'(L'+w)+e^{-w}L'(w-L')] \ , \tag{6b}$$

where $L'=L/a$.

We first note that since $L'\gg 1$, $w\leq 1$, A and B can be further simplified, and we find

$$A=(1-e^{-w})^{-1}\{1+0[L'^{-1}(2+w^2/6)]\} \ , \tag{7a}$$

$$B=(w-we^{-w})^{-1}\{1+0[L'^{-1}(2+4w^2/3)]\} \ . \tag{7b}$$

The macroscopic result, Eq. (5), will thus introduce errors of $0(1/L')$ for Peclet numbers $w\leq 1$ and is not an exact result. For $L'=15$ the exact results as determined from Eqs. (3) and (6) are $n(0)=0.0338, n(1)=0.0328$ for $w=0.1$ and $n(0)=0.0377, n(1)=0.0282$ for $w=1$. Figure 1 describes the adatom density profile across the terrace for $w=0.1$ using both the macroscopic boundary condition Eq. (5) and the microscopic boundary condition Eqs. (6). The latter shows an enhanced profile across the terrace in addition to finite, unequal values at the steps. The profiles for $0.1\leq w\leq 1.0$ are remarkably alike, and therefore we have not shown additional plots but only note that increased Peclet number leads to an increase in $n(0)$ and a decrease in $n(1)$ in agreement with expectations.

Another interesting feature is the location of the adatom density maximum on the terrace, which provides another measure of the skew produced by the step motion. Setting $n_x=0$, we find that the macroscopic result, Eq. (5), gives

$$wx=\ln[w/(1-e^{-w})] \tag{8a}$$

when $n_{Mx}=0$ and, from Eq. (3),

$$wx=\ln wA \tag{8b}$$

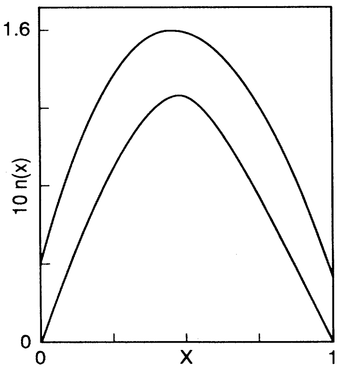

FIG. 1. Adatom density ($\times 10$) along a terrace for $L'=15$, $w=0.1$. The profiles for $0\leq w\leq 1$ are very similar. The lower curve is from Eq. (5), the macroscopic result. The upper curve is from Eqs. (3) and (6), the microscopic result.

when $n_x = 0$. Again note that when Eq. (8b) is used with Eq. (4), this is a general result for arbitrary step kinetics. For absorbing steps A is given by Eq. (6a) and approximately by Eq. (7a), the latter leading again to the macroscopic result plus a correction term. Some representative macroscopic/microscopic results are $w = 0.1, x = 0.4958/0.4948$ and $w = 1, x = 0.4586/0.4491$, with $L' = 15$. As might be anticipated, the use of symmetric boundary conditions in the macroscopic description leads to a reduction in the amount of skew produced by the moving step.

IV. CONCLUDING REMARKS

When the step kinetics are asymmetric it is possible for the adatom densities at the steps to be equal; in this case the effect of the moving step must be compensated for by a buildup on the downstream region of the terrace resulting from a reduced sticking coefficient. Let $h_1 = \beta h_0$, where β is a parameter to be determined. Then if $n(0) = n(1)$ it follows from Eqs. (3) and (4a) that $A = (1 - e^{-w})^{-1}$ and

$$\beta = [1 - e^{-w}(1+w)]/[w + e^{-w} - 1] . \tag{9}$$

The limiting value for $w \to 0$ is $\beta \to 1$, and some representative values in the range we are considering are $\beta = 0.967$ and 0.718 for $w = 0.1$ and 1.0, respectively. To relate the above result to the physical quantities S_0 and S_1, we can use Eqs. (A4b) and (A5b). As an example, consider the case where the step at $x = 0$ is absorbing, $S_0 = 1$, so that

$$\beta h_0 = \beta = h_1 = [S_1(1 - \mathrm{erf} V^*) + 2\,\mathrm{erf} V^*]/[2 - S_1(1 - \mathrm{erf} V^*)] \tag{10}$$

and

$$S_1 = (2\beta - 2\,\mathrm{erf} V^*)/(1+\beta)(1 - \mathrm{erf} V^*) . \tag{11}$$

Even in this simple case the result is a very complicated function of w. However, since $V^* \alpha w / L'$, we will have $\mathrm{erf} V^* \ll 1$ for values of w in the range we are considering, and

$$S_1 \cong 2\beta(1+\beta)^{-1} + 0(\beta\,\mathrm{erf} V^*) , \tag{12}$$

which leads to values of S_1 of 0.938 and 0.836 for $W = 0.1$ and 1.0, respectively.

Our primary purpose has been to show that the modified BCF equation describing step motion as occurs in MBE must be solved using boundary conditions that properly account for the motion-imposed asymmetry. The adatom densities at the steps must differ if the exchange kinetics are symmetric but can be equal when the downstream step is less efficient than the opposite step at capturing incident adatoms.

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APPENDIX: STEP BOUNDARY CONDITIONS

The adatom position-velocity distribution function is approximated as

$$f_i = N_i e^{-(1/2)v^2/2kT}/(2\pi kT)^{1/2} , \tag{A1}$$

where f_1 describes adatoms having velocity $v > 0$, and f_2 those having $v < 0$ in the laboratory frame; the adatom mass is taken as unity. Then

$$N = \int dv\, f = \tfrac{1}{2}(N_1 + N_2) \tag{A2a}$$

$$J = \int dv\, vf = (kT/2\pi)^{1/2}(N_1 - N_2) . \tag{A2b}$$

At the right step, X_r, which is moving with a velocity V, the density of incident and emergent adatoms are

$$N_{\mathrm{in}} = \int_V^\infty dv\, f(X_r, v) = \tfrac{1}{2} N_1 \mathrm{erfc} V^* \tag{A3a}$$

$$N_{\mathrm{out}} = \int_{-\infty}^V dv\, f(X_r, v) = \tfrac{1}{2} N_2 (1 + \mathrm{erf} V^*) , \tag{A3b}$$

with $V^* = V/(2kT)^{1/2}$. The sticking coefficient at the right step S_r (or S_1) is given as $S_r = 1 - [N_{\mathrm{out}}/N_{\mathrm{in}}]_{X = X_r}$, so that from Eqs. (A2) and (A3) and Fick's law, we get the boundary condition

$$-N_X = H_r N, \quad X = X_r, \quad \text{or} \quad -n_x = L' h_1 n, \quad x = 1 , \tag{A4a}$$

where

$$H_r = (2kT/\pi)^{1/2}[S_r(1 - \mathrm{erf} V^*) + 2\,\mathrm{erf} V^*]/[2 - S_r(1 - \mathrm{erf} V^*)] D = h_1/a . \tag{A4b}$$

Similarly, for the left step we find

$$n_x = L' h_0 n, \quad x = 0 , \tag{A5a}$$

with

$$h_0/a = [S_0(1 + \mathrm{erf} V^*) - 2\,\mathrm{erf} V^*](2kT/\pi)^{1/2}/[2 - S_0(1 + \mathrm{erf} V^*)] D . \tag{A5b}$$

Note that in the limit $w \to 0$ we recover the symmetric result $h_1 = h_0$ for $S_1 = S_0$.

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[1] W. Burton, N. Cabrera, and F. Frank, Philos. Trans. R. Soc. London A **243**, 299 (1951).

[2] A. Myers-Beaghton and D. Vvedensky, Phys. Rev. B **42**, 5544 (1990).

[3] A. Myers-Beaghton and D. Vvedensky, Phys. Rev. A **44**, 2457 (1991).

[4] V. Fuenzalida, Phys. Rev. B **44**, 10 835 (1991).

[5] S. Harris, Phys. Rev. B **47**, 10 738 (1993).

[6] Y. Tokura, H. Saito, and T. Fukui, J. Cryst. Growth **94**, 46 (1989).

[7] R. Ghez and S. Iyer, IBM J. Res. Dev. **32**, 804 (1988).

[8] This is a commonly used condition; other references could be cited.

[9] S. Harris, J. Cryst. Growth **97**, 319 (1989).

[10] S. Harris, Surf. Sci. Lett. **291**, L730 (1993).