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Onset of fast step-velocity oscillations during growth by molecular-beam epitaxy

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The kinetic equation of Burton, Cabrerra, and Frank, with an added convective term to account for step motion, is analytically investigated with the primary goal of determining the critical value of the Peclet number for which step-velocity oscillations occur. We consider the case where the adatom density vanishes at both steps, which has been previously studied by numerical analysis, and obtain a value of 3.42 for the critical Peclet number, which compares well with the numerical estimate of about 3 found earlier. We then consider the more realistic case where the down step blocks, i.e., there is a barrier to interlayer transport, and determine that in the range of Peclet numbers 0–15 no transition occurs. We provide an explanation for this and discuss some of the implications of these results relative to possible mechanisms responsible for the observed oscillations in reflective high-energy electron-diffraction intensity measurements during growth on vicinal semiconductor surfaces.

I. INTRODUCTION

The primary purpose of this paper is to provide and demonstrate a theoretical basis for predicting the onset of step-velocity oscillations that can occur during the growth of vicinal surfaces by molecular-beam epitaxy. Such oscillations have been identified as a possible mechanism responsible for the intensity oscillations observed in reflecting high-energy electron-diffraction (RHEED) studies of growing surfaces[1] in the absence of nucleation on the terraces. We will not stress this specific connection here since nucleation effects[2] are likely to be significant in this regard,[3] and we do not consider these here. An important conclusion of our paper, which also has significance for the case where nucleation is considered, is that the presence of a barrier at the down step is an important factor in determining whether a transition to oscillatory step motion will occur.

Because of the complicated nature of this problem there has been no previous analytical treatment that we are aware of; earlier work has been numerical and includes finite-difference solutions[1,2] of the generalized adatom diffusion equation incorporating step motion and simulations.[4] The approach taken here, described in Sec. II, avoids a frontal attack on the direct problem, which appears highly formidable. Instead we consider the situation where the system is allowed to evolve to a steady state under a set of conditions (miscut, temperature, and beam strength) such that a subsequent incremental increase in the beam strength leads to an immediate onset of step-velocity oscillations and also provides the basis for a linear analysis.

To implement this approach we first consider (Sec. III) the case where the adatom density vanishes at both terminating steps;[5] these are the boundary conditions used in the previous work cited above. In Ref. 2 an estimate is provided of "... about $\alpha_c=3$" (α_c is the critical Peclet number, defined in Sec. II) and we find here $\alpha_c=3.42$. We then turn in Sec. IV to the case where the down (right) step blocks. Like the first case considered, this is also a limiting case; here the down-step barrier[6] that creates the Schwoebel effect[7] is being emphasized. We find that in this case there is no transition to oscillatory step velocity for Peclet numbers in the range 0–15. This result indicates that nucleation is likely to provide the primary mechanism for observed RHEED oscillations. This point is discussed in Sec. V, which concludes this paper.

II. THEORETICAL FORMULATION

We consider growth by deposition from a beam of strength F on a vicinal surface with uniform terraces of length L having site density a^{-2}. In the absence of nucleation, and disregarding evaporation, the adatom density $n(x,t)$ satisfies a Burton-Cabrera-Frank-type equation, which we write in the coordinate system comoving with the step that has velocity $v(t)$:[1,2,4]

$$n_t=Dn_{xx}+vn_x+F,\quad 0\le x\le L\ , \tag{1}$$

where D is the diffusion coefficient. Since deposition begins at $t=0$, $n(x,0)=0$ and we must also specify boundary conditions at $x=0,L$. The step velocity $v(t)$ is given as

$$v(t)=a[Dn_x(0,t)-Dn_x(L,t)]+v(t)[n(0,t)-n(L,t)]a\ , \tag{2}$$

which is the source of difficulty in attempting to directly treat Eq. (1) by standard analytical methods.

The steady-state solution of Eq. (1) for arbitrary boundary conditions is easily found and this provides us with the basis for the strategy we employ in what follows. Before we begin, it is convenient to rewrite Eq. (1) in terms of dimensionless variables, using xL, $v_{ss}v$ ($v_{ss}=FLa$ is the steady-state step velocity), t/a^2F, and $(FL^2/D)n$ to replace the corresponding variable sin Eqs. (1) and (2) and the initial, boundary conditions so that these equations become[2]

$$n_t = 1 + n_{xx} + \alpha v n_x, \quad 0 \le x \le 1 , \tag{3}$$

$$v = [n_x(0,t) - n_x(1,t)] + \alpha v [n(0,t) - n(1,t)] , \tag{4}$$

where $\alpha = FL^2a^2/D$ is the dimensionless Peclet number.

Suppose for given F, L, and temperature (determining D) we allow the system to come to a steady state n_{ss}, v_{ss} such that a subsequent incremental increase in F, δF, producing incremental changes $\delta v, \delta n$ will initiate oscillatory step motion, i.e., δv will oscillate as a function of time. Then F, L, and D define the critical Peclet number above which the step velocity experiences a transition from strict monotone to oscillatory change to its steady-state value. We can determine the critical Peclet number α_c from the solution to Eqs. (3) and (4) with the appropriate initial/boundary conditions, where only terms linear in the incremental velocity and density are retained. Then, e.g., writing $n = n_{ss} + \delta n$, and dropping the δ's below to simplify notation, these equations become

$$n_t = f + n_{xx} + \alpha(v_{ss} n_x + v n_x), \quad 0 \le x \le 1 , \tag{5}$$

$$\begin{aligned} v = {} & [n_x(0,t) - n_x(1,t)] + \alpha v_{ss}[n(0,t) - n(1,t)] \\ & + \alpha v [n_{ss}(0,t) - n_{ss}(1,t)] , \end{aligned} \tag{6}$$

with $n(x,0) = 0$, $f = (\delta F/F)$.

The explicit solution of Eqs. (5) and (6) depends crucially on the choice of boundary conditions. Previous work[1,2,4] has considered the symmetric condition of vanishing density at the steps; as we have noted elsewhere[5] this does not imply symmetric step kinetics because the presence of the convective term implies that either the kinetics or the adatom densities must differ at the steps. We begin by considering this case, which will allow us to compare our result for α_c with that numerically estimated in previous work.[2]

III. VANISHING STEP DENSITIES

When the boundary conditions are[1,2,4]

$$n(0,t) = n(1,t) = 0 , \tag{7}$$

then the steady-state solution of Eq. (3) is

$$n_{ss} = \alpha^{-1}[(1 - e^{-\alpha x})(1 - e^{-\alpha})^{-1} - x] . \tag{8}$$

Note that the step velocity is also dimensionless and that $v_{ss} = 1$, and that for this boundary condition only $v = [n_x(0,t) - n_x(1,t)]$, i.e., the second term on the right-hand side of Eq. (4) is identically zero. The solution to Eq. (5) with Eq. (7) is

$$n = (2/\alpha) \sum_{m=1} \int_0^t dt' e^{-(t-t')(b_m^2/\alpha + \alpha/4)} \sin b_m x \int_0^1 dx' e^{-(1/2)\alpha(x-x')} \sin b_m x' [f + \alpha v n_{x'}] , \tag{9}$$

where $b_m = m\pi$. In order to obtain an explicit result we will need to approximate the above sum. Since the external parameters have been chosen so that the system will immediately commence step-velocity oscillations, it is reasonable to retain only the first two terms in the sum. Then we can determine the behavior of v by first Laplace transforming and then finding an equation for $\tilde{v}(s)$ from the expressions for $\tilde{n}(0,s)$, $\tilde{n}(1,s)$. This leads to an equation of the form

$$\tilde{v}(s) = f[A + Bs^{-1}][s^2 + Cs + D]^{-1} , \tag{10}$$

where s is the Laplace variable, A and B are constants whose values are unimportant for what follows, and C, D are also constants, given by

$$\begin{aligned} C = {} & s_1 s_2 + (2s_2/s_1)(\pi/\alpha)^2[(1 + e^{-(1/2)\alpha})(1 + e^{(1/2)\alpha}) - \alpha(1 + e^{-(1/2)\alpha})(1 - e^{-(1/2)\alpha})^{-1}] \\ & + (2s_1/s_2)(2\pi/\alpha)^2[(1 - e^{-(1/2)\alpha})(1 + e^{(1/2)\alpha}) - \alpha(1 - e^{-(1/2)\alpha})(1 + e^{-(1/2)\alpha})^{-1}] , \\ \equiv {} & s_1 s_2 + s_2[\mathrm{I}] + s_1[\mathrm{II}] , \end{aligned} \tag{11}$$

$$D = s_1 + s_2 + [\mathrm{I}] + [\mathrm{II}] , \tag{12}$$

where

$$s_i = [b_i^2/\alpha + \alpha/4], \quad i = 1,2 . \tag{13}$$

The condition for $v(t)$ to oscillate is $C^2 < 4D$, and this occurs when $\alpha = 3.42$, in reasonable agreement with the estimate given in Ref. 2 of "about $\alpha_c = 3$."

IV. BLOCKING AT THE DOWN STEP

It has been known for some time the asymmetric step kinetics provide a mechanism for step-width stability.[7] Further, it appears that in some cases the down step may actually prevent interlayer transport.[6] Thus it is probably more realistic to consider the case where the down step blocks, $n_x(1,t) = 0$, than that where the density vanishes. We might anticipate a diminished effect in this case since the down-step boundary condition will lead to a more even coverage across most of the terrace away from the up step and, in the absence of nucleation oscillatory step flow, is due to the step accelerating (decelerating) through high (low) coverage areas.

The steady-state solution to Eq. (3) for the blocking boundary condition is

$$n_{ss} = (e^{\alpha}/\alpha^2)(1 - e^{-\alpha x}) - (x/\alpha) , \tag{14}$$

and once again Eq. (9) gives the solution to Eq. (5), where now $b_m = \frac{1}{2}m\pi$ are the eigenvalues appropriate to the boundary conditions $n(0,t) = n_x(1,t) = 0$ (the eigenfunctions $\sin b_m x$ remain the same). An equation for $\tilde{v}(s)$ then follows from Eq. (6), which has the same general form as Eq. (10) with

$$C = s_1 s_2 - \alpha(e^{\alpha} - 1)^{-1}\{(b_1 - \alpha e^{-(1/2)\alpha})s_2[\mathrm{I}] + (b_2 + 2\alpha e^{-(1/2)\alpha})s_1[\mathrm{II}]\} , \quad (15)$$

$$D = s_1 + s_2 - \alpha(e^{\alpha} - 1)^{-1}\{(b_1 - \alpha e^{-(1/2)\alpha})[\mathrm{I}] + (b_2 + 2\alpha e^{-(1/2)\alpha})[\mathrm{II}]\} , \quad (16)$$

where here s_i is again given by Eq. (13) with the b_i appropriate to the blocking boundary condition and

$$[\mathrm{I}] = 2(s_1\alpha^2)^{-1}[b_1 e^{\alpha} - \alpha e^{(1/2)\alpha} - b_1] , \quad (17)$$

$$[\mathrm{II}] = 2(s_2\alpha^2)^{-1}[b_2 e^{\alpha} + \alpha e^{(1/2)\alpha} - b_2] . \quad (18)$$

We find that for $\alpha \leq 15$ the condition $C^2 < 4D$ indicating step-velocity oscillations is not satisfied and we conclude that for realistic values of α there is no transition to oscillatory step flow for the blocking condition. Thus, the effect of the blocking down step is to extinguish rather than diminish the transition to oscillatory step motion in the range of Peclet numbers of possible interest. Previous studies,[1,2,4] which focused on the symmetric case of vanishing step densities, are therefore much more limited as far as their implications than might have been expected.

V. DISCUSSION

In addition to demonstrating an approach to solving a previously intractable problem, we believe the results found here will provide guidance for future theoretical efforts towards understanding the basis of step-velocity oscillations and their role in providing a mechanism for RHEED intensity oscillations. Based on the results found here we conclude, in agreement with Ref. 2, that nucleation and subsequent incorporation of islands into the advancing step is the primary mechanism that needs to be elucidated. This requires, at the very least, that terms representing dimer formation and breakup be included in Eq. (5); however, based on our earlier work[3] we expect that these terms will include a prefactor of $\alpha^{3/2}(L/a)$ rather than the $\alpha(L/a)^2$ that has been used previously.[2] Thus the critical value of this prefactor was previously found to be $1.7\times(10^{-3})$ for $(L/a)=18$, or $\alpha_c = 1.7\times10^{-3}$ for the case of irreversible nucleation, whereas based on a corrected prefactor $\alpha_c = 3.06\times10^{-2}$. However, this assessment does not take into account the effect of the down-step barrier, and it remains to be seen what impact this might have. We conjecture that this will enhance the effect of nucleation by increasing the number of prenucleation adatoms on the terraces, leading to increased dimer density at fixed α. The difficulties in treating this problem by the method used here are twofold. First, the steady-state solution to Eq. (2) with additional terms added to account for nucleation effects is not yet known. More significant, if this solution is found, it will be necessary to solve a more complicated equation than Eq. (5), which includes a term with a nonconstant coefficient. Despite these difficulties, we believe that we have provided a theoretical framework within which this more complicated problem can be considered.

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[1] G. Petrich, P. Pukite, A. Wowchak, G. Whaley, P. Cohen, and A. Arrot, J. Cryst. Growth **95**, 23 (1989).

[2] A. Myers-Beaghton and D. Vvedensky, Phys. Rev. B **42**, 9720 (1990).

[3] We have recently shown [S. Harris, Phys. Rev. B **49**, 2967 (1993)] that the relaxation time used in Ref. 2 to describe dimer formation overestimates this effect significantly.

[4] J. Tsao, *Fundamentals of Molecular Beam Epitaxy* (Academic, San Diego, 1993), Sec. 6.3.2.

[5] The use of this boundary condition here is for purposes of comparison only, and we do not imply the same physical interpretation as in Refs. 1, 2, and 4; see S. Harris, Phys. Rev. B **48**, 8286 (1993).

[6] P. Šmilauer and D. Vvedensky, Phys. Rev. B **48**, 17 603 (1993).

[7] R. Schwoebel, J. Appl. Phys. **40**, 614 (1969).