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# Step Motion on Crystal Surfaces\*

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It is postulated that steps on crystal surfaces capture atoms diffusing on the surface with certain probabilities and, in addition, that the capture probability depends on the direction from which adsorbed atoms approach the step. A general solution for the time-dependent step distribution is obtained in terms of these probabilities and an arbitrary initial distribution of an infinite sequence of parallel steps. It is shown that coalescence of steps or stabilization of step spacings can occur as a consequence of assuming that capture probabilities are directionally dependent. Some of the implications of the theoretical model are related to the growth of real crystal surfaces.

## INTRODUCTION

ONE of the characteristic morphological features of growing crystal surfaces is the presence of step systems in which the spacing between succeeding steps changes in some slowly varying manner. These step systems frequently exhibit typical step heights which are considerably greater than that of a monolayer. The development of step systems in which the steps are of multiple height has been treated in a number of ways. Cabrera<sup>1</sup> has discussed development of macroscopic steps as a means of reducing the surface energy of the crystal. Amelinckx, Bontinck, and Dekeyser<sup>2</sup> have shown that a sequence of monatomic steps around screw dislocations can bunch into macroscopic steps for certain values of the dissolution or growth parameters. Frank,<sup>3</sup> Cabrera and Vermilyea,<sup>4</sup> and Chernov<sup>5</sup> have discussed step bunching in terms of shock waves of steps using a continuum theory for step motion. Mullins and Hirth<sup>6</sup> have considered a system of differential equations for step motion and have discussed several aspects of step bunching or facet formation for a system consisting of a finite number of steps. A number of investigators have also suggested that heterogeneities on the surface, such as impurities, may slow the growth of a step to such an extent that succeeding steps overtake it and produce a macroscopic step.

In each of the previous publications dealing with the kinetics of step motion it has been assumed that steps behave like ideal sinks. That is, it has been assumed that atoms diffusing on the surface are incorporated into steps with unit probability. This assumption stems

from a hypothesis of step behavior postulated by Burton, Cabrera, and Frank<sup>7</sup> in their theory of crystal growth. It has recently been shown, however, that atoms diffusing on surfaces of the parent crystal may be either reflected from, or reversibly captured at steps. Ehrlich and Hudda,<sup>8</sup> using field ion microscopy, have shown that atoms diffusing on the surface of an emitter tip may be reflected at the step boundary. Schwoebel,<sup>9</sup> using replication techniques and electron microscopy, has shown that single-crystal surfaces of gold growing from the vapor phase develop characteristic growth centers and oriented step systems depending on the crystallography of the substrate. The latter observations indicate that steps of different crystallographic orientations incorporate diffusing atoms into the advancing step face with different probabilities. It is concluded from such experimental evidence that steps do not, in general, capture diffusing atoms with unit probability.

In this paper we consider some of the consequences of assuming that a step incorporates a diffusing atom into itself with a certain probability. Furthermore, we assume that the direction from which an atom approaches the step determines the associated probability of capture. For simplicity, a model is considered in which only two probabilities are involved and these are associated with the two sides from which atoms may approach a step. For certain relative values of these probabilities it is shown that the step spacing can be stabilized, and, for other relative values it is shown that steps can coalesce with one another to form steps of greater than the minimum height.

In the next section we (1) indicate why a step may incorporate diffusing atoms into it with a probability which depends on the direction of approach, (2) develop and solve a system of differential equations for

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<sup>1</sup> N. Cabrera, *J. Chem. Phys.* **21**, 1111 (1953).

<sup>2</sup> S. Amelinckx, W. Bontinck, and W. Dekeyser, *Phil. Mag.* **2**, 1264 (1957).

<sup>3</sup> F. C. Frank, in *Growth and Perfection of Crystals*, R. H. Doremus, B. W. Roberts, and D. Turnbull, Eds. (John Wiley & Sons, Inc., New York, 1958), p. 411.

<sup>4</sup> N. Cabrera and D. A. Vermilyea, in Ref. 3, p. 393.

<sup>5</sup> A. A. Chernov, *Soviet Phys.—Usp.* **4**, 116 (1961).

<sup>6</sup> W. W. Mullins and J. P. Hirth, *J. Phys. Chem. Solids* **24**, 1391 (1963).

<sup>7</sup> W. K. Burton, N. Cabrera, and F. C. Frank, *Phil. Trans. Roy. Soc. London* **243A**, 299 (1951).

<sup>8</sup> G. Ehrlich and F. G. Hudda, *J. Chem. Phys.* **44**, 1039 (1966).

<sup>9</sup> R. L. Schwoebel (unpublished).

the step spacings in an infinite sequence of steps, and (3) describe the morphological evolution of some step systems which have particularly simple distributions at  $t=0$ . In the final section some of the implications of this growth model in the growth of single crystals are discussed.

### THE GROWTH MODEL

The interdependence of the capture probability for an atom diffusing near a step, and the relative direction of the atom from the step, can be illustrated by considering a cross section of the surface as shown in Fig. 1. The associated potential energy for a diffusing atom might appear as shown in the lower portion of Fig. 1 in which the two modifications in the potential (at A and B) arise from: (1) the decreased coordination for a diffusing adsorbed atom at A, and (2) the increased coordination at B. The potential may actually be quite different from that illustrated in Fig. 1, but atoms adsorbed in the regions to the left or right of the step in Fig. 1 will have, in general, different probabilities of moving into position c at the step. Although there are several conceivable motions of an atom at positions a and b (reflection from the step, transmission across the step, diffusion parallel to the step, etc.), we are concerned only with the probability that atoms at a or b move into position c at the step. The probability of an adsorbed atom at a (Fig. 1) moving into the step is called  $\gamma_-$  and the probability for an atom adsorbed at b moving into the step is designated  $\gamma_+$ . It is shown that inequalities of the  $\gamma$ 's result in step growth dependent primarily on one of the two areas bounding the step on which adsorbed atoms are diffusing. Accordingly, both the spacing between steps (i.e., the areas feeding the advancing step face) and the value of  $\gamma_+$  and  $\gamma_-$  determine the step velocities. General

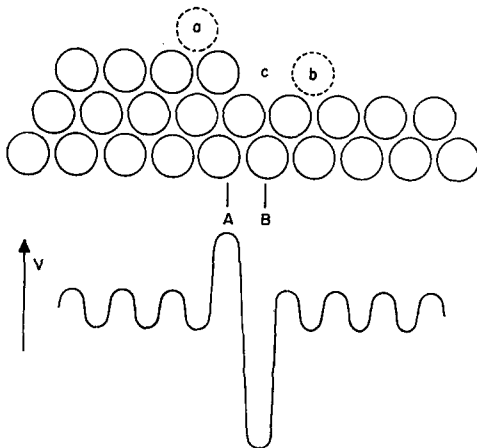


FIG. 1. The cross section of a monatomic step in a surface and the hypothetical potential associated with the diffusion of an atom over such a surface. Two major modifications of the potential may occur at A and B due to the decreased and increased coordination between a surface diffusing atom and the substrate, respectively. In terms of this hypothetical potential, it might be expected that steps advance primarily through the incorporation of atoms diffusing in the region to the right of the step.

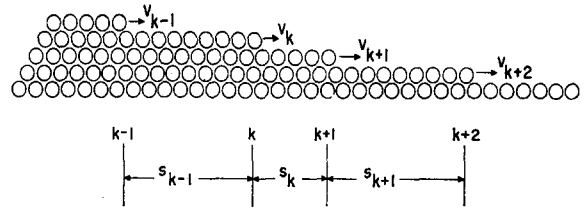


FIG. 2. A cross section of the step model which initially consists of an infinite sequence of parallel steps of minimum height descending to the right.

solutions for the step spacings as a function of time are obtained. A few simple distributions of step spacings at  $t=0$  are considered to illustrate how step coalescence or the stabilization of step spacing may occur as a consequence of the inequality of  $\gamma_+$  and  $\gamma_-$ .

A simple model of the surface is considered which initially consists of an infinite sequence of parallel steps of minimum height descending to the right, Fig. 2. Growth of this hypothetical surface proceeds by deposition of atoms onto the areas between steps and subsequent surface diffusion of these adsorbed atoms to steps. It is initially assumed that the diffusion length of an adsorbed atom is large in comparison to the step spacing at  $t=0$ , and that the motion of steps can be described in terms of a one-dimensional diffusion problem. The steps (Fig. 2) are indexed  $\dots, k-1, k, k+1, \dots$  and the associated step spacings between the  $k$  and  $k+1$  step is  $s_k$ , etc.

The rate of change of the spacing  $\dot{s}_k$  between steps  $k$  and  $k+1$  is

$$\dot{s}_k = v_{k+1} - v_k, \quad (1)$$

where the  $v_k$ 's are the step velocities. The step velocities can be expressed in terms of the spacing (which is directly related to the feed area) on each side of the step. For the case in which the diffusion length of the adsorbed atom is much greater than the step spacing, these are of the form

$$v_k = C(s_k\gamma_+ + s_{k-1}\gamma_-). \quad (2)$$

In this expression,  $C$  is assumed to be a constant of the step motion and  $s_k$  and  $s_{k-1}$  are the spacings on either side of the  $k$ th step (Fig. 2). The expression for  $\dot{s}_k$  is then

$$\dot{s}_k = C[\gamma_+s_{k+1} + (\gamma_- - \gamma_+)s_k - \gamma_-s_{k-1}]. \quad (3)$$

It is convenient to define  $Y_k$  as

$$Y_k = s_k \exp[(\gamma_+ - \gamma_-)Ct], \quad (4)$$

since the derivatives of  $s_k$  can then be expressed in terms of the  $Y$ 's as

$$\begin{aligned} \dot{Y}_k &= C(\gamma_+Y_{k+1} - \gamma_-Y_{k-1}), \\ \ddot{Y}_k &= C^2(\gamma_+^2Y_{k+2} - 2\gamma_+\gamma_-Y_k + \gamma_-^2Y_{k-2}), \\ \dddot{Y}_k &= C^3(\gamma_+^3Y_{k+3} - 3\gamma_+^2\gamma_-Y_{k+1} \\ &\quad + 3\gamma_+\gamma_-^2Y_{k-1} - \gamma_-^3Y_{k-3}), \\ &\vdots \\ Y_k^{(n)} &= C^n \sum_{m=0}^n \frac{(-1)^m n!}{m!(n-m)!} \gamma_+^{n-m} \gamma_-^m Y_{k+n-2m}. \end{aligned} \quad (5)$$

The general solution for  $Y_k(t)$  can be expressed as

$$Y_k(t) = \sum_{n=0}^{\infty} \frac{Y_k^{(n)}(0)t^n}{n!} \\ = \sum_{n=0}^{\infty} \frac{(Ct)^n}{n!} \sum_{m=0}^n \frac{(-1)^m n!}{m!(n-m)!} \gamma_+^{n-m} \gamma_-^m s_{k+n-2m}(0). \quad (6)$$

This expression for  $Y_k(t)$  can be written in terms of Bessel functions after some rearrangement of the right-hand side. Equation (6) can first be written as

$$Y_k(t) = \sum_{m=0}^{\infty} \sum_{n=m}^{\infty} \frac{(-1)^m (Ct)^n}{m!(n-m)!} \gamma_+^{n-m} \gamma_-^m s_{k+n-2m}(0). \quad (7)$$

Making the substitution  $p = n - 2m$ , Eq. (7) becomes

$$Y_k(t) = \sum_{m=0}^{\infty} \sum_{p=-m}^{\infty} \frac{(-1)^m (Ct\sqrt{\gamma_+\gamma_-})^{p+2m}}{m!(p+m)!} \left(\frac{\gamma_+}{\gamma_-}\right)^{p/2} s_{k+p}(0). \quad (8)$$

Again, rewriting the sums in Eq. (8),  $Y_k(t)$  can be expressed as

$$Y_k(t) = \sum_{p=0}^{\infty} \sum_{m=0}^{\infty} \frac{(-1)^m (Ct\sqrt{\gamma_+\gamma_-})^{p+2m}}{m!(p+m)!} \left(\frac{\gamma_+}{\gamma_-}\right)^{p/2} s_{k+p}(0) \\ + \sum_{p=-1}^{-\infty} \sum_{m=-p}^{\infty} \frac{(-1)^m (Ct\sqrt{\gamma_+\gamma_-})^{p+2m}}{m!(p+m)!} \left(\frac{\gamma_+}{\gamma_-}\right)^{p/2} s_{k+p}(0). \quad (9)$$

Using the definition of Bessel functions, Eq. (9) can be written as<sup>10</sup>

$$Y_k(t) = \sum_{p=0}^{\infty} J_p(2Ct\sqrt{\gamma_+\gamma_-}) \left(\frac{\gamma_+}{\gamma_-}\right)^{p/2} s_{k+p}(0) \\ + \sum_{p=-1}^{-\infty} J_p(2Ct\sqrt{\gamma_+\gamma_-}) \left(\frac{\gamma_+}{\gamma_-}\right)^{p/2} s_{k+p}(0), \quad (10)$$

or

$$Y_k(t) = \sum_{-\infty}^{\infty} J_p(2Ct\sqrt{\gamma_+\gamma_-}) \left(\frac{\gamma_+}{\gamma_-}\right)^{p/2} s_{k+p}(0). \quad (11)$$

The general solution for  $s_k(t)$  is, using Eqs. (4) and (11),

$$s_k(t) = [\exp(\gamma_- - \gamma_+)Ct] \\ \times \sum_{-\infty}^{\infty} J_p[2Ct(\gamma_+\gamma_-)^{1/2}] \left(\frac{\gamma_+}{\gamma_-}\right)^{p/2} s_{k+p}(0). \quad (12)$$

This is the general solution for the spacing  $s_k(t)$  associated with the  $k$ th step in terms of the  $\gamma$ 's and the initial step spacing at  $t=0$ . Two features of this solution indicate some of the differences that can be expected by assuming that the  $\gamma$ 's may be unequal. First, the

<sup>10</sup> For these and other Bessel function relations, see: E. T. Whittaker and G. N. Watson, *Modern Analysis* (Cambridge University Press, New York, 1958).

exponential term involving  $\gamma_- - \gamma_+$  indicates that important changes in the distribution will occur with time. Second, it can be noted that terms in the sum are weighted by factors of  $(\gamma_+/\gamma_-)^{p/2}$ . If  $\gamma_+ > \gamma_-$ , steps ahead of the  $k$ th step (i.e.,  $k+1, k+2, \dots$ ) are of principal importance in determining the spacing associated with the  $k$ th step. The steps ( $k-1, k-2, \dots$ ) behind the  $k$ th step are more influential if  $\gamma_- > \gamma_+$ . An equivalent statement is that the areas ahead or behind the  $k$ th step are more important to step growth, depending on whether  $\gamma_+$  or  $\gamma_-$  is larger, respectively. We now consider three forms of the step spacing at  $t=0$ : (1) a uniformly spaced sequence of steps; (2) a sequence of steps in which the spacing varies sinusoidally; and (3) a sequence of step spacings which are monotonically increasing or decreasing. Nonuniform distributions are of principal interest since these are more representative of the real surface. It is seen in cases (2) and (3) that differences in  $\gamma_+$  and  $\gamma_-$  can produce particular modifications of the initial surface structure as can certain classes of initial step distributions.

#### Case 1: $s_k(0) = L$ , a Constant, for all $k$

First, we consider that the steps are equally spaced a distance  $L$  from one another at  $t=0$ . For this case, the general solution (7) can be obtained using the generating function for Bessel functions,

$$\exp\left[\frac{z}{2}\left(t - \frac{1}{t}\right)\right] = \sum_{n=-\infty}^{\infty} t^n J_n(z). \quad (13)$$

The expression for  $s_k(t)$  is then

$$s_k(t) = L[\exp(\gamma_- - \gamma_+)Ct] \\ \times \exp\{Ct(\gamma_+\gamma_-)^{1/2}[(\gamma_+/\gamma_-)^{1/2} - (\gamma_-/\gamma_+)^{1/2}]\} = L. \quad (14)$$

Therefore, a uniform distribution of step spacings at  $t=0$  is independent of both time and the values of  $\gamma_+$  and  $\gamma_-$ .

#### Case 2: $s_k(0) = L(1 + A \cos Kk)$ for $K, L = \text{Constants}$

Second, a sinusoidal distribution of step spacings with an average spacing of  $L$  at  $t=0$  is discussed. In this case, the general solution (7) can be simplified considerably by again using the generating function Eq. (13) for Bessel functions. The result is that

$$s_k(t) = L(1 + A\{\exp[Ct(\gamma_- - \gamma_+)(1 - \cos K)] \\ \times \{\cos[Kk + Ct(\gamma_+ + \gamma_-) \sin K]\}). \quad (15)$$

To demonstrate the form of the solution for this case, let us consider that  $K = \pi$ , i.e., that the initial distribution of step spacings is alternately  $L(1+A)$ ,  $L(1-A)$ ,  $L(1+A)$ ,  $L(1-A)$ ,  $\dots$ . The expression for  $s_k(t)$  is then

$$s_k(t) = L(1 + A\{\exp[2Ct(\gamma_- - \gamma_+)]\} \cos k\pi). \quad (16)$$

For this special case we can note that:

(a) if  $\gamma_- < \gamma_+$ , all step spacings approach a uniform spacing  $L$  which is the average spacing at  $t=0$ ;

(b) if  $\gamma_- = \gamma_+$ , there is no change of the initial distribution of step spacings;

(c) if  $\gamma_- > \gamma_+$ , the step spacings associated with even-numbered steps grow exponentially in time while the spacings associated with the remaining steps decrease as growth proceeds. This motion would eventually lead to coalescence of adjacent steps resulting in the formation of a sequence of uniformly spaced steps separated by a distance  $2L$ . Each step would be of *double* height at time  $t_c$ .

$$t_c = \ln A / 2C(\gamma_+ - \gamma_-), \quad (17)$$

where  $t_c$  is the characteristic time for coalescence in this case.

The example chosen for case 2 ( $K = \pi$ ) is a particularly simple one in which the change of the initial distribution of step spacings with time may be clearly seen. Although the changes of step spacing occurring in systems with a more complicated initial distribution of  $s_k$ 's cannot be simply stated, some of the general features of the evolving step system can be noted. These general features are, for the case in which the initial step distribution is nonuniform:

(1) If  $\gamma_- < \gamma_+$ , an arbitrary initial distribution of step spacings will tend toward a sequence of uniformly spaced steps.

(2) If  $\gamma_- = \gamma_+$ , step spacings will oscillate in time as growth of the surface proceeds. The general solution for  $s_k(t)$  then represents an enumeration of normal modes for the oscillation of step spacings in analogy to spacings in a linear lattice in which only nearest-neighbor interactions are assumed. Clearly, steps can group into bunches or waves in this case, just as acoustic waves of various wavelengths can form in a linear lattice.

(3) If  $\gamma_- > \gamma_+$ , some step coalescence will occur if the initial distribution is not uniform.

### Case 3: $s_k(0) = Lr^k$ , for Constant $L$ and $r$

If  $r > 1$ , the step system is characterized by spacings which are successively larger. Accordingly, if  $r < 1$ , successive steps are closer to one another. Using the generating function Eq. (13) we can solve for  $s_k(t)$  as

$$s_k(t) = Lr^k \exp\{Ct[\gamma_-(1-1/r) + \gamma_+(r-1)]\}.$$

For this case, it may be noted that:

(a) if  $r > 1$ , the spacing of successive steps increases exponentially with time, regardless of the relative values of  $\gamma_+$  and  $\gamma_-$ ;

(b) if  $r < 1$ , successive step spacings decrease exponentially in time, regardless of  $\gamma_+$  and  $\gamma_-$ , and a steep front of closely spaced steps is formed.

The results for each of these three cases are valid when the surface diffusion length is long in comparison to the

step spacing. If, for example, step coalescence is occurring because  $\gamma_- > \gamma_+$ , the average step spacing on the surface is increasing. This kind of description of the evolution of a coalescing step system is appropriate until this relation between the diffusion length and the average step spacing is no longer valid.

## DISCUSSION

In the preceding section we have attempted to illustrate a mechanism by which morphological changes of the step system may occur on idealized surfaces. The examples of initial step spacing distributions were selected to emphasize two principal features of these hypothetical step systems.

(1) The relative values of  $\gamma_+$  and  $\gamma_-$  may influence the development of a step system either by producing more uniform spacings or by causing steps to coalesce into steps of multiple height.

(2) Certain types of initial step distributions can dominate the subsequent morphological development of the step system, independent of the values of  $\gamma_+$  and  $\gamma_-$ . Such step distributions can occur locally; however, it is believed the  $\gamma$  values will determine the essential structural features in the evolution of a real crystal surface.

This model departs from some of the earlier theories since it allows for the possibility of step coalescence rather than only a temporary grouping of steps as a wave packet. The possibility of monatomic steps grouping into packets (forming a "step" of multiple height) is a special case ( $\gamma_- = \gamma_+$ ) of the theory presented here. On phenomenological grounds one might expect that  $\gamma_+$  and  $\gamma_-$  would be different on real surfaces and that their values would depend on the crystallographic details of the step. If this is the case, a growing crystal surface could develop some steps of multiple height as a natural consequence of the capture characteristics of the step. For the same reason other steps (e.g., of different crystallographic orientation) might never develop comparable vertical dimensions because of different relative values of  $\gamma_+$  and  $\gamma_-$ . This idea is consistent with results of morphological studies of crystal surfaces growing from the vapor phase in which the observations indicate that steps of certain crystallographic orientations can develop vertical dimensions which are considerably larger than steps of other orientations.<sup>9</sup> It is possible, of course, to envision other mechanisms of macroscopic step development which are related to the trapping or capture of adsorbed atoms near or at a step. An example of an alternate mechanism is one in which adsorbed atoms are trapped in position d [Fig. 3(a)] at the upper boundary of the step. If subsequent growth proceeds to the left from an atom at d [Fig. 3(a)], a new step of monolayer height has been generated and the original step is now of multiple height.

Although there have been several studies of the macroscopic morphology of growing crystal surfaces,

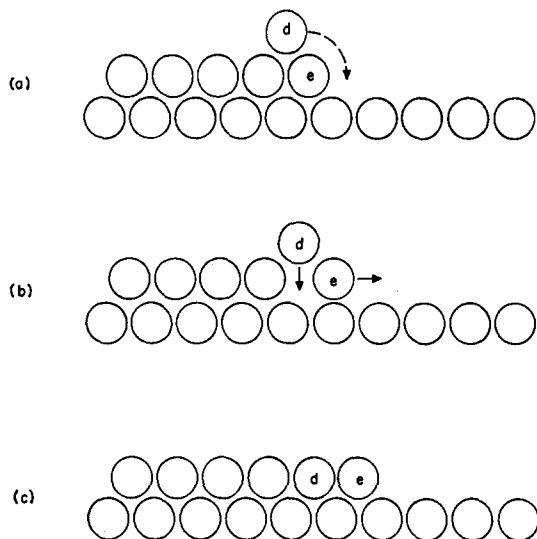


FIG. 3. Schematic representation of correlated motion of atoms near a step resulting in growth of the step. Motion of the atom labeled *d* may be directly to the step position [as designated by the dashed arrow in 3(a)] in which the activation energy is related to the diminished coordination required for such motion. However, it is possible that for steps and surfaces with certain crystallographic features, correlated motion of atoms near the step 3(b) could produce the same result with a different characteristic activation energy.

very limited information is available concerning the distribution of steps with heights equivalent to ten monolayers or less on such surfaces. It is interesting to consider that it may be possible to observe such structure not only through use of direct observational techniques such as electron microscopy, but perhaps also by means of diffraction phenomena. If the step spacing is of sufficient regularity throughout the width of an impinging electron beam (for example, in LEED apparatus), effects of diffraction from a superlattice might be observed which are due solely to a morphological characteristic of the surface, i.e., a regularly spaced sequence of steps.

We have mentioned in earlier paragraphs that the relative values of  $\gamma_+$  and  $\gamma_-$  may depend on the crystallographic details associated with the step, such as the crystal plane on which the step is located and the crystallographic direction of the step on that plane. It is tempting to conclude that  $\gamma_-$  will generally be smaller than  $\gamma_+$  because of the diminished coordination that is required for an atom to pass from position *d* [Fig. 3(a)] over position *e* directly to the step site. We believe,

however, that there may be a variety of ways in which an atom at position *d* may contribute to the advancement of the step face. An example of this is shown schematically in Fig. 3(b) in which the correlated motion of *two* atoms results in the advance of the step by one atomic dimension, Fig. 3(c). What is suggested by this example is that there may be a variety of correlated motions through which the step face may be advanced and these mechanisms may quite reasonably be connected to the crystallographic details surrounding the step. Accordingly, general conclusions regarding the relative values of  $\gamma_-$  and  $\gamma_+$  are perhaps premature and considerably more detailed considerations are required. The definition of  $\gamma_-$  and  $\gamma_+$  must therefore be broadened to include the possibility that adsorbed atoms near the step may contribute to the advance of a step face in indirect ways.

The similarities between the structural development of this highly idealized model of the surface and a real surface are, of course, very limited. The model which we have considered is valid when the characteristic diffusion length of an adsorbed atom is larger than the step spacing and for a time equal to that of first coalescence. Behavior of the system following first coalescence has not been discussed. Although any step distribution at  $t=0$  may be considered in the formalism, the general solution [Eq. (12)] is valid only for the special case in which the steps are initially all the same height and the same sign. As soon as coalescence occurs, a new set of equations must be considered. To generalize this kind of treatment to include a random distribution of steps of arbitrary height and sign at  $t=0$ , one must consider that the constant of the step motion  $C$  is specific to each step and hence must be indexed accordingly. Such a generalization complicates the resulting equations considerably. It must be recognized in such an extension that transport on the step face must be considered and that steps of multiple height may be characterized by new values of  $\gamma_+$  and  $\gamma_-$  which are distinguished from those for steps of minimum height. The values of  $\gamma_+$  and  $\gamma_-$  may also be dependent on such parameters as the kinetics of the growth process, adsorbed impurities at or near steps, etc. Considerations of this kind would, however, be particularly germane to relating the ideas expressed in this paper to the concepts of crystal evaporation, annealing, thermal etching, and other processes which may depend in part on surface transport.