Formation of macroscopic steps at vicinal surfaces during crystal growth

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The evolution of the macroscopic relief of a vicinal surface of a growing (or evaporating) crystal is analyzed. An equation is derived to describe the formation and decay of macroscopic steps as a result of nonlinear effects of a diffusion interaction of moving elementary steps. Depending on the extent of supersaturation, this equation may reduce to either a Burgers equation or a Korteweg–de Vries equation. Some particular exact solutions of these equations which are of most interest for applications are analyzed.

1. Experiments have shown that under certain conditions during the growth (or evaporation) of a crystal a characteristic stepped relief will form on thermodynamically stable vicinal surfaces.[1,2] Analysis shows that the observed formation of macroscopic steps may be the result of the excitation of shock waves in the density of moving elementary steps.[3]

In this letter we analyze nonlinear effects during the diffusion interaction of moving elementary steps on a vicinal surface, with the goal of finding a systematic description of the morphology of the macroscopic steps which are formed as a function of the external parameters.

2. We consider a thermodynamically stable vicinal surface of the $(1K0)$ type, tilted at a small angle θ_0 from a close-packed singular plane. If a surface of this sort were ideally planar at the macroscopic level, it would consist of elementary steps of height a separated by terraces of uniform length $l_0 \approx a/\theta_0$. In other words, it would consist of equidistant echelon of parallel elementary steps with a density $\rho_0 = l_0^{-1}$. A real surface differs from an ideal surface in that there are macroscopic irregularities on it; i.e., the local density of steps, $\rho(X)$, oscillates around a mean value[1] ρ_0.

Under crystal growth conditions, each of the elementary steps moves at a velocity which is determined by the diffusion fluxes of adatoms from the adjacent terraces:

$$v_i = D_a \left(\frac{\partial \xi_i}{\partial x} \bigg|_{+0} - \frac{\partial \xi_i}{\partial x} \bigg|_{-0} \right), \tag{1}$$

where D_a is the self-diffusion coefficient of the adatoms, and $\xi_i(x)$ is the distribution of the concentration of adatoms on terrace i, which is determined from the equation[3]

$$\lambda_s^2 \frac{\partial^2 \xi_i}{\partial x^2} - (\xi_i - \xi_s) = 0 \tag{2}$$

under the boundary conditions

$$\xi_i\Big|_{x=0} = \xi_i^0; \qquad \xi_i\Big|_{x=l_i} = \xi_{i+1}^0 \equiv \xi_i^0 + \Delta, \tag{3}$$

where $\lambda_s = (D_a\tau_a)^{1/2}$ is the mean diffusion path length of an adatom over its lifetime τ_a on the surface, and ξ_s is the concentration of adatoms which is at equilibrium with the supersaturated vapor around the crystal. The dependence of the equilibrium adatom concentration ξ^0 on the curvature $K = a(\partial\rho/\partial X)$ is incorporated phenomenologically in (3):

$$\xi^0(K) = \xi_0 (1 + K\gamma\omega /kT),$$

where γ is the surface tension, ω is the atomic volume, and ξ_0 is the equilibrium concentration of adatoms on an ideally plane surface. The increment (Δ) in the equilibrium adatom concentration per terrace is

$$\Delta = \frac{\delta\xi^0}{\delta X} l = \frac{\alpha}{\rho} \frac{\partial^2\rho}{\partial X^2} , \tag{4}$$

where $\alpha = \gamma\omega a\xi_0/kT$.

Calculating v_i from (1) and (2), using (3) and (4), and transforming to a dimensionless step density $\eta(X) = \lambda_s [\rho(X) - \rho_0]$ (under the assumption $|\eta| \ll \lambda_s\rho_0$), we can work from the continuity equation

$$\frac{\partial\rho}{\partial t} + \frac{\partial}{\partial X} (\rho v) = 0$$

to construct an equation describing the evolution of an echelon of steps of variable density:

$$\frac{\partial\eta}{\partial\tau} - u_0 \frac{\partial\eta}{\partial Y} - B\eta \frac{\partial\eta}{\partial Y} - \kappa \frac{\partial^2\eta}{\partial Y^2} + C \frac{\partial^3\eta}{\partial Y^3} = 0, \tag{5}$$

where $\tau = v_0 t /\lambda_s$ is the dimensionless time, $v_0 = 2D_a\xi_0/\lambda_s$ is the normalized velocity of an isolated elementary step, $Y = X /\lambda_s$ is the dimensionless coordinate, $u_0 = \epsilon_0^3 \sigma_s / 3$, σ_s is the supersaturation in the vapor phase, $\epsilon_0 = 1/2\lambda_s\rho_0$ is the mean dimensionless length of a terrace, $B = 4\epsilon_0^2 \sigma_s$, $\kappa = \epsilon_0^2 \sigma_s + C$, and $C = \gamma\omega a/2kT\lambda_s^2$. We have thus reduced the problem at the microscopic scale, of the diffusion of atoms over terraces, to a problem at the macroscopic scale, involving the evolution of the density of steps.

The qualitative picture of the events depends on a competition between the nonlinear effects which are determined by the third term in (5), on the one hand, and dissipative effects (the fourth term) or dispersive effects (the last term), on the other.

3. Since the coefficient κ in (5) contains two terms, it is clear that if the supersaturation in the vapor, σ_s, is sufficiently high,

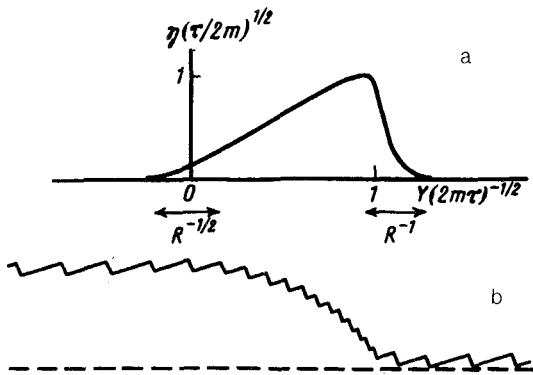$$\sigma_s \gg \gamma\omega a\rho_0^2 /kT , \tag{6}$$

FIG. 1. a—Distribution of the density of elementary steps; b—normalized profile of a macroscopic step ($R = Bm/2\kappa$ is the Reynolds number).

we find the well-known Burgers equation[4]

$$\frac{\partial \eta}{\partial \tau} - u_0 \frac{\partial \eta}{\partial Y} - B\eta \frac{\partial \eta}{\partial Y} - \kappa \frac{\partial^2 \eta}{\partial Y^2} = 0, \tag{7}$$

for which we can derive exact expressions for the evolution of a wide range of initial conditions $\eta(Y, \tau = 0) \equiv f(Y)$. Here we will interpret only the two solutions which we regard as the most interesting.

A. Under the initial condition $f(Y) = m\delta(Y)$, which determines the presence of a macroscopic step with a height of m elementary steps on the vicinal surface of interest, the solution of the Burgers equation is a triangular wave (Fig. 1). It moves at a velocity $V = (2m\tau)^{1/2}$ with respect to the echelon in the direction opposite the direction in which the steps move. The evolution of the initial macroscopic step during the growth occurs in such a way that the step density distribution remains constant in the coordinates of Fig. 1. This statement means that a step moving at a velocity V slowly (in proportion to $\tau^{1/2}$) "spreads out" over time.

B. In the case of periodic initial condition $f(Y) = \eta_0 \sin(2\pi Y/L)$, we find a sawtooth wave (Fig. 2) with a discontinuity amplitude $\Delta\eta$ which varies over time, as a result of the evolution of $\eta(Y, \tau)$. This result means that macroscopic steps appear on a surface which is essentially smooth in its initial state. A discontinuity in the density of elementary steps arises at the time $\tau_1 = L/2\pi\eta_0$, and its height $\Delta\eta$ increases over
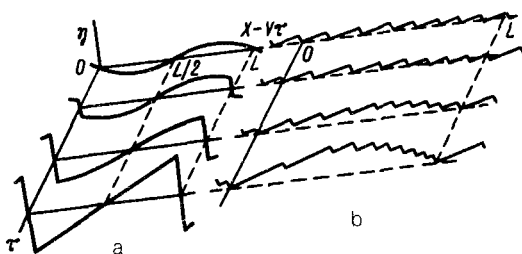

FIG. 2. a—Appearance of a discontinuity in the density of elementary steps; b—surface profile of the crystal, for the case of periodic initial conditions.

time, goes through a maximum at $\tau_2 = L/4\eta_0$ and then falls off, so we have $\Delta\eta \approx L/\tau$.

If $\sigma_s \ll 2\gamma a\rho_0^2/kT$, the evolution of the relief is determined by a competition between the nonlinear effects associated with dissipative and dispersive properties, which are related exclusively to capillary forces. The evolution of a perturbation with a typical length $\lambda \gg \lambda_s$ is again described by a Burgers equation, but if $\lambda \ll \lambda_s$ we instead find a Korteweg-de Vries equation

$$\frac{\partial \eta}{\partial \tau} - u_0 \frac{\partial \eta}{\partial Y} - B\eta \frac{\partial \eta}{\partial Y} + C \frac{\partial^3 \eta}{\partial Y^3} = 0. \tag{8}$$

In this case the structure of the effects is qualitatively different. The evolution of spatially localized initial conditions can be described by choosing solitary-wave solutions which correspond to a perturbation region which is moving along the surface of the crystal:

$$\eta(Y, \tau) = \frac{3V/B}{\cosh^2\{(V/C)^{1/2}(Y + V\tau + u_0\tau)\}}. \tag{9}$$

In contrast with the preceding case, neither the shape of the perturbation nor its propagation velocity V decays as time elapses.

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[1]We are using two length scales: the macroscopic length scale X, a length element of which, δX, contains some number of elementary steps $\delta n = \rho\delta X$; and a microscopic length scale x, whose length element δx is significantly smaller than the length of a step.

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[4]G. B. Whitham, Linear and Nonlinear Waves, Wiley-Interscience, New York, 1974.

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