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Kinematic Multiplication of Elementary Steps on a Crystal Surface

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The dynamics of elementary steps on an atomically smooth crystal–liquid interface and, in particular, the process of collisions of steps differing in sign are considered. It is shown that, along with the conventional annihilation of steps in such collisions, both the overthrow of steps to the neighboring row with the formation of a new atomic layer (passage) and the reflection of steps from each other can take place under certain conditions. The overthrow of steps gives a qualitatively new mechanism of the growth of facets in the absence of renewable sources such as grown-in dislocations. Under these conditions, the growth kinetics of a crystal with atomically smooth facets changes substantially. In particular, the processes considered above may form a basis for physical mechanisms of unconventional growth regimes observed for helium crystals at low temperatures. © *2003 MAIK "Nauka/Interperiodica".*

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It is well known that the growth of a crystal with atomically smooth facets can proceed due to either continuously acting sources of elementary steps such as screw dislocations and Frank–Read sources or two-dimensional nucleation. The impossibility of crossings is an essential property of steps, which is commonly not questioned. For steps of the same sign, this means the impossibility of one atomic layer to hang over another, that is, the impossibility of a configuration with a high excessive energy. From the same energy considerations, it is clear that, upon coming in contact, two steps of different signs annihilate in the contact region with the formation of a bridge (Fig. 1a). The step noncrossing property was used as the basis in classical works on the theory of crystal growth [1].

The property of steps indicated above is undoubtedly retained as long as all the processes with the participation of steps are sufficiently slow, so that each step section at each instant of time is in a local equilibrium and the kinetic energy of a step can be neglected. In other words, the corresponding relaxation time must be small in comparison with the "collision time" w/V, where V is the relative velocity of steps and w is the characteristic width of a step, which equals the interatomic distance by the order of magnitude. This condition can easily be violated in the case of atomically smooth facets of a helium crystal at temperature tending to zero, when the relaxation time increases without limit and the step velocities can be very high up to the sound velocity [2]. It is natural to suggest that the colliding steps of different signs in this case will be able to pass inertially one over the other forming a new atomic layer (see Fig. 1b) or to reflect from each other. We will show in this brief communication that this actually takes place under certain conditions. Here it is necessary to note that this idea in itself is analogous to the idea of the "kinematic multiplication" of dislocations in

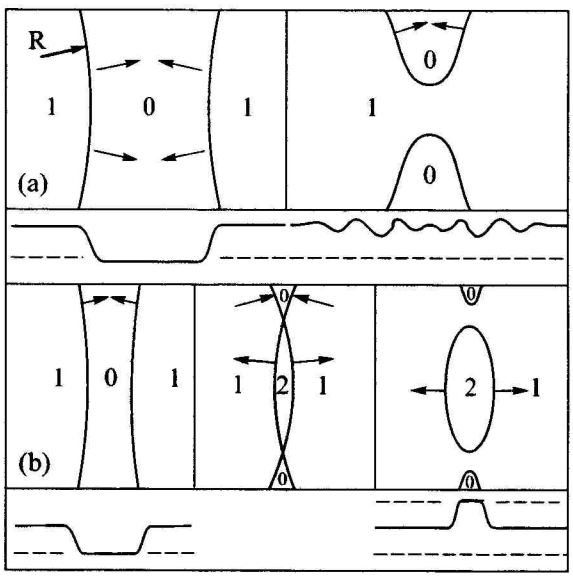

Fig. 1. (a) Collision of steps in a quasi-static case. (b) Passage of steps one over the other at high velocities and large relaxation times. A nucleus of a new layer is formed at this place as a result of the collision.

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the crystal bulk, which was proposed by Frank [3] even before the discovery of Frank–Read sources.

We will describe the structure and dynamics of steps using the so-called weak-coupling approximation, which is widely used in the theory of phase transitions associated with the initiation of faceting in helium crystals (see, for example, [4]). In this approximation, the effective periodic potential retaining the surface in the vicinity of certain equilibrium positions is small in comparison with the surface energy because of strong fluctuations. Correspondingly, the energy of a step, that is, the transition region between two neighboring equilibrium positions of the surface, is also small, and the width of this region is conversely large in comparison with the interatomic distance. This allows one to introduce a continuous variable $\zeta(\mathbf{r})$ corresponding to a local displacement of the surface averaged over the fluctuations. Consider the case of extremely low temperatures. Then the dissipation, which accompanies the motion of steps, and the external supersaturation, which is necessary for its maintenance, are small, and the corresponding terms in the equations of motion can be neglected. In addition, we will consider both the liquid and the crystal incompressible. Then a displacement of the surface is associated only with crystallization or melting, and the total energy is a sum of the surface and kinetic energies. The surface energy minus an inessential constant can be written as

$$H_{\text{surf}} = \int\frac{\alpha}{2}(\nabla\zeta)^2 d^2\mathbf{r} + \int U_0\left(1-\cos\frac{2\pi\zeta}{a}\right)d^2\mathbf{r}, \quad (1)$$

where the first term takes into account a change in the surface area, the second term corresponds to the contribution of the effective potential, a is the interplanar distance, and α is the energy of unit surface area (here, we make no distinction between the surface energy and stiffness).

The kinetic energy in this case is the kinetic energy of the liquid, whose motion is due to the displacement of the surface;[1] with regard to the conservation of mass in crystallization, we obtain

$$H_{\text{kin}} = \frac{1}{4\pi}\frac{(\rho_s-\rho_l)^2}{\rho_l}\int\frac{\partial\zeta(t,\mathbf{r})}{\partial t}\frac{\partial\zeta(t,\mathbf{r}')}{\partial t}\frac{d^2\mathbf{r}d^2\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}, \quad (2)$$

where ρ_s and ρ_l are the densities of solid and liquid helium. The corresponding equation of motion takes the form

$$\frac{1}{2\pi}\int\frac{\partial^2\varphi}{\partial t^2}\frac{d^2\mathbf{r}}{|\mathbf{r}-\mathbf{r}'|} - \Delta\varphi + \sin(\varphi) = 0, \quad (3)$$

[1] The additional contribution due to the rearrangement of atoms on passing from the liquid to the crystalline state [5] is relatively small and does not affect the qualitative conclusions.

where dimensionless coordinates x and y are measured in units ξ and time, in units τ

$$\xi = \sqrt{\frac{\alpha a^2}{4\pi^2 U_0}}, \quad \tau = \sqrt{\frac{\Delta\rho^2}{\rho_l}\frac{a^2}{4\pi^2 U_0}}\xi,$$
$$\varphi = \frac{2\pi\zeta}{a}, \quad \Delta\rho = \rho_s - \rho_l, \quad (4)$$

and U_0, in turn, can be expressed through the energy β of the unit length of immobile step

$$U_0 = \frac{\pi^2}{16}\frac{(\beta/a)^2}{\alpha}. \quad (5)$$

Let us also give here numerical values for the (0001) facet of a ^{4}He crystal: $\alpha = 0.25$ erg/cm^2 [6–8], $\beta/a = 0.014$ erg/cm^2 [8], and the other parameters

$$U_0 \approx 1.5\times10^{-3}\alpha, \quad \xi \sim 4a, \quad \tau \sim 5\times10^{-12}\text{ s},$$
$$V_0 = \xi/\tau \sim 2.8\times10^4\text{ cm/s} < c \approx 3.6\times10^4\text{ cm/s}, \quad (6)$$

where c is the speed of sound in the liquid.

The real steps on a crystal surface are always somewhat curved (see Fig. 1), and the radius of curvature R commonly exceeds or, at least, coincides by the order of magnitude with the radius of the critical nucleus R_c. Because $R_c \gg \xi$ (otherwise, the probability of conventional two-dimensional nucleation would be high; note also that the value of R_c is inversely proportional to the external supersaturation, which we assume to be small), the inequality $R \gg \xi$ is also fulfilled, so that steps in collisions first come in contact in a region $R^* \sim \sqrt{R\xi}$, which is small in comparison with R, where they can be considered rectilinear and parallel to each other. Therefore, first we must analyze the one-dimensional case, in which instead of Eq. (3) we have

$$\frac{1}{\pi}\int\ln\left(\frac{R^*}{|x'-x|}\right)\frac{\partial^2\varphi}{\partial t^2}dx' = \frac{\partial^2\varphi}{\partial x^2} - \sin\varphi. \quad (7)$$

This equation was solved numerically in the following sequence. First, the function $\varphi(x - Vt)$, which described the stationary shape of a single step moving with a prescribed velocity V measured in units V_0, was defined. Note that, at $V = 0$, this shape coincides with the shape of a single kink of the sine-Gordon equation and deviates more and more from this shape with increasing V. At $V > 1$, the stationary shape in the form of a simple kink loses its stability. The question of which configurations are stable in this case still remains unexplored.

At the second stage, the function corresponding to the stationary shape of two steps of different signs moving toward each other with velocities V_1 and V_2 was used as the initial condition for the solution of the problem of collision between such two steps. It was found that, depending on the values of initial velocities, three

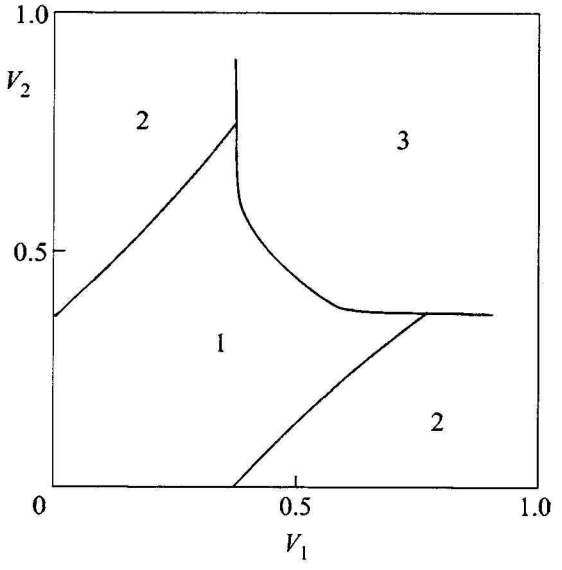

Fig. 2. Schematic diagram of collision processes of steps moving with different velocities of counter motion: regions correspond to (1) annihilation of steps, (2) reflection, and (3) passage with the creation of a new atomic layer.

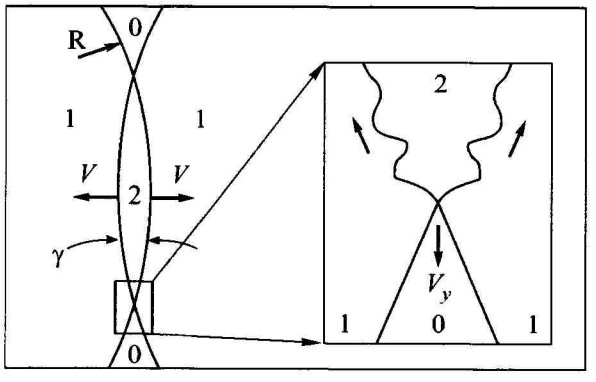

Fig. 3. Generation of excitations at the crossing point of steps and their propagation along the boundary of the newly formed layer.

qualitatively different results could be obtained: the passage of step over one another with "overthrow" to the neighboring row, annihilation, and reflection. All three processes are accompanied by the emergence of "ripplons" of higher or lower amplitudes. Regions corresponding to each of these processes are indicated in the schematic diagram in Fig. 2.

The further evolution of the system is substantially different in these three cases. Case (3), when a nucleus of a new atomic layer confined by two intersecting arcs of steps is formed (Fig. 1b), is of most interest. Generally speaking, it should be expected that an instability leading eventually to the formation of a bridge, as is shown in Fig. 1b, will develop in the crossing region. However, the terms in the total equation of motion (3) responsible for this instability are small at the initial instant of time by virtue of the smallness of the crossing angle γ; that is, the corresponding lifetime is large. At the same time, the result of the action of perturbations is not accumulated with time, because the crossing point itself moves along the Y axis with a velocity $V_y \cong 2V/\gamma$, which is faster than the velocity of the propagation of perturbations along the step ($V_{exc} \leq 1$, see Fig. 3). Therefore, the formation of a bridge becomes possible only at $\gamma \sim 1$; that is, when the size of the nucleus of the new layer l is on the order of R, and thus $l > R_c$. We can see that actually no additional restriction for the stable growth of the nucleus of the new layer arises in comparison with the schematic diagram in Fig. 2.

Note in conclusion that, with regard to the phenomenon considered above, the theory of the growth of atomically smooth crystal facets at low temperatures requires substantial corrections. In particular, it is possible that the proposed growth mechanism provides the basis for the so-called "burstlike" growth of dislocation-free helium crystals observed experimentally [2], which defies explanation within the known growth mechanisms.

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