Crystallization Waves on an Atomically Smooth Surface

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For the atomically smooth surfaces (faces) of a quantum crystal, the possibility of the presence of traveling waves similar to crystallization waves observed on an atomically rough surface is demonstrated. However, unlike the latter wave type, the waves on atomically smooth surfaces are nonlinear even when their amplitudes are small. The propagation velocity of such waves is determined as a function of their amplitude and wavelength.

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It is generally agreed that crystallization waves, i.e., weakly damped oscillations that arise because of the periodic melting and crystallization at the surface of a quantum crystal contacting the superfluid liquid phase [1, 2], are only possible at an atomically rough boundary, but not at an atomically smooth one (i.e., on a crystal face). The point is that, for the existence of crystallization waves, it is necessary that the melting and crystallization processes be virtually nondissipative. This condition is satisfied at sufficiently low temperatures for the atomically rough surface of a helium crystal. The rough surface is characterized by a fast increase in the kinetic growth coefficient with decreasing temperature in contrast to the smooth surface, for which the growth coefficient is exponentially small in supersaturation at any temperature (a review concerning this problem and other problems of the kinetics and thermodynamics of helium crystal surfaces can be found in [3]).

A vicinal surface, i.e., a surface close in its orientation to one of the crystal faces, remains in the atomically rough state up to the temperature of the corresponding roughening transition. Therefore, the crystallization waves on this surface do not basically differ from the crystallization waves on other rough surfaces. At the same time, the vicinal surface can be considered as an echelon of elementary steps of the same "sign." Hence, the growth and melting processes on the vicinal surface can be interpreted as a result of the motion of elementary steps. From this point of view, a wave with the wave vector perpendicular to the step line is nothing but the step density wave (see Fig. 1). Since all of the steps are of the same sign, the amplitude of the surface slopes in this wave is bounded by the angle θ between the unperturbed vicinal surface and the initial face. It should also be noted that, for the surface to be considered as a stepped one, it is necessary that the average distance between the steps be small compared to the effective step width ξ , which can be widely different for different faces and, in particular, can be much greater than the step height *a* [3]. In other words, the value of the angle θ itself is bounded by the condition $\theta \ll a/\xi$.

Now, the question arises as to whether the waves simultaneously containing steps of both signs are possible. In this respect, the case of an initial surface containing no steps at all, i.e., the case of an atomically smooth initial surface, seems to be of most interest (Fig. 2). The answer to this question is the purpose of the present paper.

First of all, we note that the propagation of the wave shown in Fig. 2 is only associated with the motion of the existing steps rather than with the formation of new steps (or their disappearance). Therefore, at sufficiently low temperatures, at which the mobility of the steps is high, such a process should be virtually nondissipative. We stress that this consideration refers to traveling waves rather than standing ones. However, it is still unclear whether waves of this type are possible. In addition, for the experiment, it is important to know how these waves can be excited.



Fig. 1. Schematic representation of a crystallization wave on a vicinal surface. The ratio between the height of the elementary step and the wave amplitude is artificially increased for the sake of illustration.

We limit our consideration to plane waves $\zeta(x, t)$ with a macroscopic amplitude ζ_0 . We assume that the liquid and the crystal are incompressible and the amplitude is sufficiently small compared to the wavelength, so that $|\zeta_x| \ll a/\xi$. Then, the equation of motion for the surface can be represented in the form (compare [4])

$$\frac{(\rho_c - \rho_l)^2}{\pi \rho_l} \int \ln \left(\frac{R}{|x' - x|} \right) \ddot{\zeta}(x') dx' - \tilde{\alpha}(\zeta_x) \zeta_{xx} = 0, \quad (1)$$

where ρ_c and ρ_l are the densities of the crystal and the liquid, respectively; $\tilde{\alpha}$ is the surface stiffness; and *R* is the surface size in the direction perpendicular to the *x* axis.

Equation (1) represents the local (for a given *x*) equilibrium condition for the surface. According to the aforementioned, this condition can be considered satisfied only within the sloping (rough) parts of the surface $(\zeta_x \neq 0)$. Within atomically smooth areas $(\zeta_x = 0)$, the condition $\dot{\zeta} = 0$ is satisfied instead of Eq. (1). Hence, the integration in Eq. (1) is actually performed over the same region within which the solution is sought, i.e., in the region where $\zeta_x \neq 0$. In view of the condition $|\zeta_x| \ll a/\xi$, we discuss here the vicinal surfaces, for which, according to the existing concepts (see [3]), we can assume that $\tilde{\alpha} = \gamma |\zeta_y|$.

We seek the solution to Eq. (1) in the form of a traveling wave $\zeta(x, t) = \zeta(x - Vt)$ with a wavelength λ . Let the wave have the simplest form (as in Fig. 2), where sequential half-waves differ only in the sign of ζ_x (i.e., $\zeta_x(x + \lambda/2) = -\zeta_x(x)$ while, within each single half-wave, ζ_x does not change sign. Then, in Eq. (1), we can perform the preliminary summation over individual halfwaves and the integration within one half-wave with $\zeta_x \ge 0$)

$$\frac{\left(\rho_{c}-\rho_{l}\right)^{2}V^{2}}{\pi\rho_{l}}\int_{-\lambda/4}^{\lambda/4}\ln\tan\frac{\pi}{\lambda}|x'-x|\zeta_{x'x'}dx'+\gamma\zeta_{x}\zeta_{xx}=0.$$
 (2)

This equation cannot be solved analytically (except for one specific case, see below). Therefore, we seek the solution numerically, by the variational method. The corresponding functional is nothing but the energy (per unit area of the surface) in the frame of reference moving with a velocity V:

$$E = \frac{(\rho_c - \rho_l)^2 V^2}{\pi \rho_l \lambda} \int_{-\lambda/4}^{\lambda/4} \ln \tan \frac{\pi}{\lambda} |x' - x| \zeta_x \zeta_{x'} dx dx' + \frac{\gamma}{3\lambda} \int_{-\lambda/4}^{\lambda/4} \zeta_x^3 dx.$$
(3)

As already mentioned, the desired function ζ_x can be nonzero not within the whole interval ($-\lambda/4$, $\lambda/4$), but

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Fig. 2. Crystallization wave on an atomically smooth surface.

only within its part ($-\lambda \varepsilon/4$, $\lambda \varepsilon/4$), where the value of ε is to be determined. In addition, at a given velocity V and a given amplitude ζ_0 , the wavelength λ is also determined by the minimum condition for functional (3). In fact, the variation can be conveniently performed by representing Eq. (3) in the form

$$F = \frac{1}{\nu} \int_{-1}^{1} \ln \tan \frac{\pi \varepsilon}{4} |y - y'| f(y) f(y') dy dy' + \frac{1}{\nu^{3} \varepsilon^{2}} \int_{-1}^{1} f^{3} dy, \quad E = \frac{128}{3} \frac{\gamma \zeta_{0}^{3}}{\lambda_{0}^{3}} F,$$
(4)

where $v = \lambda/\lambda_0$, $\lambda_0^2 = \frac{32\pi}{3} \frac{\gamma \rho_l \zeta_0}{(\rho_c - \rho_l)^2 V^2}$, and the function

$$f = \frac{\lambda^{\varepsilon}}{8\zeta_0} \zeta_x$$
 is normalized by the condition $\int_{-1}^{1} f \, dy = 1$.

Functional (4) reaches its minimum value $F_{\min} = -0.893$ at $\varepsilon = 0.612$, v = 1.221, and f(y) of the form

$$f(y) = \frac{2}{\pi} \sqrt{1 - y^2} \sum_{n} C_n U_n(y),$$
 (5)

where U_n are the Chebyshev polynomials of the second kind with coefficients rapidly decreasing as *n* increases: $C_0 = 1$, $C_2 = 9.3 \times 10^{-4}$, $C_4 = 1.8 \times 10^{-5}$, $C_6 < 5 \times 10^{-7}$, and $C_1 = C_3 = C_5 = 0$. We note the additional symmetry of this solution: the function *f* is an even function. As ε and v depart from the point of the aforementioned minimum, the value of the functional *F* increases rapidly, which, presumably, precludes the presence of other minima. However, we cannot completely exclude the possibility of the existence of asymmetric solutions, because (as one can easily show) the violation of symmetry leads to stepwise changes in the coefficients C_n (as in the case of a first-order phase transition).

Thus, the solution found above corresponds to the wave shown in Fig. 2, in which plane regions alternate with curved ones. This wave is stable to small perturbations of its form and of the parameters λ and ϵ ; it propagates with a velocity depending on the amplitude. The dispersion law for such a wave can be represented as

$$\omega^{2} = V^{2}k^{2} = \frac{8}{3\pi} \frac{\rho_{l}\gamma}{(\rho_{c} - \rho_{l})^{2}} v^{2}k^{4}\zeta_{0}, \qquad (6)$$

where $k = 2\pi/\lambda$ is the wave number. From the comparison with conventional crystallization waves, for which

$$\omega^2 = \frac{\rho_l \alpha}{\left(\rho_c - \rho_l\right)^2} k^3, \tag{7}$$

we see that dispersion law (6) corresponds to the effective surface stiffness $\tilde{\alpha}_{eff} = \frac{8}{3\pi} \gamma v^2 k \zeta_0$. Here, it should be noted that, throughout this paper, we ignore the presence of the field of gravity. In doing so, we assume that both the wavelength and the wave amplitude are small compared to the effective capillary length determined as $l_{eff} = \sqrt{\gamma/(\rho_c - \rho_l)g}$ (for the (0001) face of the ⁴He crystal, $l_{eff} \sim 1 \text{ mm [3]}$). This requirement together with the conditions formulated earlier determines the domain of applicability of the results obtained.

Now, we consider the particular case where $\varepsilon \longrightarrow 0$ while the product εv remains finite. In this case, Eq. (2) actually describes a solitary half-wave (a kink) of height $2\zeta_0$ and length $\Delta = \varepsilon v \lambda_0/2$, because the interaction with other kinks separated by distances that are multiples of $v \lambda_0/2$ can be ignored. Instead of *F*, the functional to be varied in this case is

$$F_{1} = \iint_{-1}^{1} \ln \varepsilon |y - y'| f(y) f(y') dy dy' + \frac{1}{\varepsilon^{2} v^{2}} \int_{-1}^{1} f^{3} dy, \quad (8)$$

which reaches its minimum at

$$f(y) = \frac{2}{\pi} \sqrt{1 - y^2}, \quad \varepsilon v = \frac{\sqrt{6}}{\pi}.$$
 (9)

The stability of this solution can be verified by representing a small perturbation $\delta \zeta$ (normalized by $2\zeta_0$) in the form of a series expansion in Chebyshev polynomials by analogy with Eq. (5). The corresponding variation of F_1 is an essentially positive quantity:

$$\delta F_1 = \frac{\pi^2}{2} \sum_n n(n+1) C_n^2.$$
 (10)

The necessary integrals of the Chebyshev polynomials can be found, e.g., in [5].

As in the case of periodic waves, one can easily verify that solution (9) satisfies the necessary conditions in wide ranges of velocities, amplitudes, and lengths Δ , which are related by the formula

$$V^{2} = \frac{16}{\pi} \frac{\rho_{l} \gamma}{\left(\rho_{c} - \rho_{l}\right)^{2} \Delta^{2}}.$$
 (11)

It is quite probable that the initial equation (1) also has other solutions stable to small perturbations, e.g., solutions corresponding to different combinations of solitary waves or periodic wave configurations more complicated than the one considered above. The investigation of all these possibilities, as well as various nonstationary processes, goes beyond the scope of this paper.

The excitation of the waves under study at the surface of a real helium crystal is basically possible by using the same methods as those used for the excitation of ordinary crystallization waves, for example, by an electric field [2, 3]. However, an important distinctive feature should be taken into account: in the case under study, the excitation of crystallization waves is of a threshold character, because it is associated with the growth and/or melting of the atomically smooth surface. The threshold magnitude (the difference between the chemical potentials of the crystal and the liquid) can be estimated as $\delta \mu \approx 2\beta/Ra$, where β and α are the energy per unit length and the height of the elementary step, respectively, and R is the critical nucleus size. The electric field E gives rise to $\delta \mu = (\epsilon_c - \epsilon_l)E^2/8\pi$, where $\epsilon_c - \epsilon_l$ is the difference between the dielectric constants of the crystal and the liquid. Then, the threshold electric field strength is

$$E_{\rm tr} \approx \sqrt{\frac{16\pi\beta}{(\epsilon_c - \epsilon_l)Ra}}.$$
 (12)

For $\beta/a = 0.014 \text{ erg/cm}^2$ (the value corresponding to the (0001) face of the ⁴He crystal) and $R = 10^{-3}$ cm, we obtain $E_{\text{tr}} \approx 10^5$ V/cm, which is quite attainable in a real experiment [2, 3].

Earlier, few observations of solitary waves at the faces of ⁴He [6] and ³He [7] crystals in the absence of an electric field were reported in the literature, but these observations were made after shaking the experimental cell or in the course of the rapid growth or melting of the crystals. The experimental conditions of all of these observations were far from being stationary, which does not allow us to interpret them as an experimental corroboration of the existence of waves discussed in this paper.

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