Hydrodynamic instability during non-uniform growth of a helium crystal

S. N. Burmistrov, L. B. Dubovskii, and V. L. Tsymbalenko
Institute of Superconductivity and Solid State Physics, Kurchatov Institute, 123182 Moscow, Russia

We analyze an analog of the hydrodynamic Rayleigh-Taylor instability for the liquid-solid phase interface under non-uniform growth of the solid phase. The development of the instability starts on conditions of an accelerated interface growth and if the magnitude of acceleration exceeds some critical value. The plane and spherical shapes of the interface are considered. The observation of the instability can be expected for helium crystals in the course of their abnormal fast growth.

PACS numbers: 81.10.h, 64.70.Dv, 67.40.Hf

Introduction. The fast growth kinetics of the crystal-superfluid helium interface results in appearing the instabilities similar to those inherent in the interfaces between fluid media. For instance, it is shown that the crystal surface is unstable in the steady tangential flow of a fluid [1, 2], demonstrating thus the Kelvin-Helmholtz type of instability. The phenomenon is observed qualitatively as a distortion of the crystal surface in the fluid jet [3]. The onset of such instability occurs when the harmonics increasing exponentially in time appear in the spectrum of crystallization waves.

It is known after [2] that the wave spectrum does not vary in the case of the normal incidence of the fluid onto the surface. Hence the growth of a crystal should not lead to an instability of its surface. However, in the experiment on the free growth of the crystal initiated at the needle point immersed into the liquid bulk [4, 5] one has observed an instability at the initial stage of the abnormal fast growth. After nucleation the crystal grows first with the clear kinetic prism-shaped faceting. Then, if the initial overpressure at which the crystal is nucleated is sufficiently large, the crystal after 0.1–0.2 ms takes the shape close to the spherical one with a sizeable ripple on the surface, see Fig. 1. The magnitude of the threshold overpressure lies within 6–8 mbar and the threshold of the instability does not depend upon the temperature within errors [6].

The attempts to explain this phenomenon within the known theoretical works have been unfruitful [6]. The essential point, which differs the experiment from the cases considered theoretically, is unsteady growth conditions. Under such large overpressures the crystal grows in the oscillating regime with the typical oscillation period ~0.4 ms and reaches the size ~1 mm for the first half-cycle. The linear velocity for the motion of the crystal boundary runs up to ~10 m/s. The interface-directed velocity of the liquid phase is one order of the magnitude as smaller due to difference in the densities of the liquid and solid phases. During the first half-cycle the crystal growth rate varies from maximum to zero one. This gives an estimate ~10⁵ m/s² for the acceleration of the interface and ~10⁴ m/s² for the fluid beside the crystal.

Another essential point here is a direction of acceleration which is opposite to the velocity of the crystal growth. In the non-inertial reference frame related to the interface segment this results in the acceleration directed from the crystal to the liquid. Since the crystal density is larger than the liquid one, the situation proves to be looking like the normal Rayleigh-Taylor instability [7, 8] for the interface between two classical immiscible fluids of various densities. The interface is unstable or stable according to whether the acceleration is directed from the lighter to the heavier fluid or vice versa.

However, the fast growth kinetics of a helium crystal in the abnormal state gives rise to a series of qualitative features. The classical Rayleigh-Taylor instability develops at the fixed volume of each phase. In contrast, the impetuous growth of the liquid-crystal interface can result in varying the interface profile due to phase conversion. In addition, besides the kinetic liquid and interface surface energies it is necessary to involve the energy of phase conversion depending on the imbalance from the phase equilibrium. Thus, in spite of superficial resemblance such instability has specific features which differ it from the familiar Rayleigh-Taylor instability.

The variation of the surface shape of a crystal at its non-uniform growth has been studied only in the experiments on abnormal crystal growth [5]. The classical instability of the crystal-liquid helium interface in the field of gravity is observed by Demaria, Lewellen, and Dahm [9]. In these experiments a cell in which the solid and liquid phases occupy initially the lower and upper halves, respectively, is inverted mechanically by 180°. After in-

FIG. 1: The growth of a crystal at 0.47 K and initial overpressure 5.2 mbar. The left frame corresponds to 0.19 ms after the crystal nucleation. The right frame is taken at 80 ms after the crystal nucleation at the pressure close to the phase equilibrium pressure. The vertical size of the frames is 2.4 mm.
version the crystal starts to melt, the interface descending along the walls and ascending in the centre of a cell. The authors gave only the schematic description of the process without concrete spatial scale and time reference points over technical failures with determining the interface position from the filming of the process. Unfortunately, the temperatures at which the experiments are performed are not indicated with the exception of the crystal growth temperature 1.1 K and maximum one 1.7 K. The question about the effect of roughening transitions upon stability of the crystal facet in the field of gravity has remained open. We note only the following. The stability observed for the shape of the crystal grown at the needle point \[9\] with its lower facet under condition favorable for developing the Rayleigh-Taylor instability evidences that the acceleration of gravity did not result in any instability of the atomically smooth interface for the sizes not exceeding 10 mm at least.

In the present work we give a theory of the phase interface instability for the case of the non-uniform interface growth rate. A comparison is presented between the interface instability for the case of the non-uniform inter-

The growth kinetics of the crystal surface. Let the plane liquid-solid interface grow at the rate \(V = \dot{L}(t)\) and regions \(z > L(t)\) and \(z < L(t)\) be occupied with the superfluid and the solid \(4\)He, respectively. We consider stability of the growing interface with respect to its small distortions from the plane shape, i.e., interface \(z\)-coordinate is taken as \(Z = L(t) + \zeta(x, y, t)\). Assuming the crystal growth rates to be small compared with the first and second sound velocities, we consider the hydrodynamics of a superfluid in the approximation of incompressible liquid and constancy of the entropy density. In this case \[9\] the equations for the normal and superfluid motions can be separated and the potentials of the normal and superfluid velocities satisfy, respectively, \(\nabla^2 \phi_n = 0\) and \(\nabla^2 \phi_s = 0\). Thus, the solutions with wave vector \(q = (q_x, q_y)\) parallel to the surface can be represented as

\[
\phi_n = u_n(t)z + A_n(t) \exp(iqr - qz) \\
\phi_s = u_s(t)z + A_s(t) \exp(iqr - qz),
\]

where \(r = (x, y)\) and velocities \(u_s\) and \(u_n\) correspond to those of the undisturbed motion. The pressure in the fluid is a sum of pressures \(P = P_n + P_s\) and

\[
P_s &= P_s(\infty) - \rho_s(\phi_s + (\nabla \phi_s)^2/2) + \rho_s g(L(\infty) - z) \\
P_n &= P_n(\infty) - \rho_n(\phi_n + (\nabla \phi_n)^2/2) + \rho_n g(L(\infty) - z).
\]

Let us employ the boundary conditions used in the theory of crystalization waves and crystal growth kinetics. From the continuity of the mass flow we have at the interface \(z = Z(x, y, t)\)

\[
\dot{j}_n = \rho_n V_n \nu + \rho_s V_s \nu = (\rho - \rho') \dot{Z}.
\]

Here \(\nu\) is the normal to the surface.

To simplify the consideration, further on we neglect shear components \(\sigma_{ij}\) for the stress tensor \(\sigma_{ik}\) in the solid phase. In other words, we suppose the stress tensor to be diagonal, i.e., \(\sigma_{ik} = -P \delta_{ik}\). Then the solid looks like a liquid under pressure \(P'\).

From the continuity of the momentum flux density and involving the surface tension and interfacial curvature, we arrive at

\[
P + \rho_n(\nu - \dot{Z} \nu) + \rho_s(\nu - \dot{Z} \nu)^2 - (P' + \rho' \dot{Z}^2) = \\
\gamma_{ik} \frac{\partial Z^2}{\partial x_i \partial x_k} = 0,
\]

where \(\gamma_{ik} = \alpha \delta_{ik} + \frac{\partial \alpha^2}{\partial \varphi_i \partial \varphi_k}\) is the surface stiffness tensor expressed in terms of surface tension \(\alpha\) and derivatives over angle \(\varphi\) between the normal to the displaced surface and the axis \(z\). The quantities with dash are referred to the solid phase.

For the normal velocity according to work \[10\], we suppose the sticking of the normal component to the surface as for a viscid fluid

\[
V_n \nu = \dot{Z}.
\]

The growth rate of the solid phase is described by the kinetic growth coefficient \(K\) and connected with the difference in chemical potentials per unit mass as usual

\[
\dot{Z} = K \left[ \mu + \frac{(\nu - \dot{Z} \nu)^2}{2} - \left( \mu' + \frac{\dot{Z}^2}{2} \right) \right].
\]

Then we have for the difference in the chemical potentials and temperature \[9\]

\[
\frac{\mu - \mu'}{\rho} = \\
= \sigma (T - T(\infty)) + \frac{P - P_s}{P_n(\infty) - P_s(\infty)} \frac{(\nu - \nu_s)^2}{(\nu - \nu_s) P_s(\infty)} - \frac{P' - P_s}{\rho_s(\infty)},
\]

where \(\sigma\) is the entropy and the quantities with index \(\infty\) stand for the magnitudes taken far from the interface. Pressure \(P_c\) denotes the equilibrium pressure at which the phase transition takes place.

Solving a set of the boundary conditions, one finds that the undisturbed motion of the plane interface at rate \(V = \dot{L}(t)\) obeys

\[
V \frac{\rho'}{\rho} = \dot{\rho}' - \frac{\rho}{\rho} \left[ \Delta P + \rho g(L(\infty) - L) \right] + \rho_{el} \left( \dot{V} L + \frac{V^2}{2} \right).
\]

Here \(\rho_{el}\) is the effective density of the interface

\[
\rho_{el} = \rho_n + (\rho' - \rho) \frac{s^2}{\rho}
\]

and \(\Delta P = P(\infty) - P_c\) is the overpressure. The equation for the disturbed motion in the linear approximation reads

\[
\rho_{el} \frac{\ddot{\zeta}}{q} + \frac{\rho'}{K} \dot{\zeta} + [\gamma_{ik} q_i q_k + (\rho' - \rho) g - \rho_{el} V] \zeta = 0.
\]
For the uniform growth of the interface $\dot{V} = 0$, the equation goes over into the familiar dispersion relation for the crystallization wave spectrum \[ 10 \ 12. \]

**The instability.** Let us consider the stability of the flat-shaped interface with respect to small perturbations $\zeta \sim \exp(\lambda t)$ for the uniformly accelerated growth of the crystal. The root with $\text{Re}\, \lambda(q) > 0$ corresponds to instability. For the acceleration exceeding the threshold one $\dot{V}_c = (\rho' - \rho)/\rho_{ct}$, the interfacial distortion will increase for the wave vectors satisfying $\gamma_{ct} q_k < \rho_{ct} (\rho' - \rho) g$. Note that $\dot{V}_c$ does not depend on the growth coefficient, i.e., on the dissipative properties of the interface, and is positive. The latter corresponds to the case when the interfacial acceleration is directed to the fluid. Thus, the stability due to non-uniform growth of the plane interface appears only for the accelerated growth of a crystal.

The value $q_0$ corresponding to the maximum magnitude $\text{Re}\, \lambda(q)$ meets the shortest time for the development of the instability which will be characterized by the spatial scale $2\pi/q_0$. The value $q_0$ can be found from the equation

$$ q_0^2 = \frac{g^2}{3} - \frac{\sqrt{2}}{3} \frac{\rho'}{\gamma_{ct} \rho K} \frac{q_0^{3/2}}{3}, $$

where $q_0$ is the value related to the upper bounds of instability according to $\gamma q_0^2 = \rho_{ct} \dot{V} - (\rho' - \rho)$. For the large magnitudes of the growth coefficient or large acceleration while $K^4 \dot{V} \gg \rho' / \gamma_{ct}^3$, values $q_0$ and $\lambda(q_0)$ are equal approximately to

$$ q_0 = \frac{q_0}{\sqrt{3}} \left( 1 - \frac{\rho'}{\gamma_{ct} \rho_0^{3/2}} \right), $$

$$ \lambda_0 = \left( \frac{2\gamma}{3\sqrt{3}} \frac{q_0}{\rho_{ct}} \right)^{1/2} - \frac{q_0}{2\sqrt{3}} \frac{\rho'}{\rho_{ct} K}. $$

In the opposite limit $K^4 \dot{V} \ll \rho' / \gamma_{ct}^3$ one has roughly

$$ q_0 = \left( \frac{\gamma_{ct} \rho_{ct} K^2}{\rho'} \right)^{1/3}, $$

$$ \lambda_0 = \frac{K}{\rho'} \gamma_{ct}^2. $$

On the whole, values $q_0$ and $\lambda_0$ decrease as the kinetic growth coefficient reduces or dissipation of the interface enhances. From the experimental point of view this may require the crystal surface of sufficiently large sizes $d > 2\pi/q_0$ and the large time of supporting the accelerated growth $t > 1/\lambda_0$ in order to realize the interfacial instability at the uniformly accelerated growth.

**The spherical geometry.** In the experiment a crystal nucleates at the needle point and then grows free. The anisotropy of the kinetic growth coefficient is not large. A ratio of the maximum to minimum growth rate does not exceed 2-3. Let us consider the stability of the spherical shape of the growing crystal. For simplicity, we assume the isotropy of the surface tension $\alpha$. As it follows from the estimates given in the Introduction, the acceleration of gravity is negligibly small as compared with the typical magnitudes of the interfacial acceleration.

The equation of the interface is given as $r = R_s(t, \Omega) = R(t) + \zeta(t, \Omega)$ where $\zeta = \zeta(t) Y_l(\Omega)$ is a fluctuation of the interface expanded in the spherical harmonics of degree $l = 0, 1, 2 \ldots$. We seek for the velocity potentials of the normal $v_n = \nabla \phi_n$ and superfluid $v_s = \nabla \phi_s$ motions in the form

$$ \phi_s = -u_s R^2/r + A_l Y_l/r^{l+1}, $$

$$ \phi_n = -u_n R^2/r + B_l Y_l/r^{l+1}. $$

Using the same boundary conditions as above, we find the velocities of the undisturbed flow of the liquid phase

$$ u_s = \dot{R} (\rho_s - \rho') / \rho_s, \quad u_n = \dot{R}, $$

and coefficients $A_l$ and $B_l$ describing the disturbed motion of the interface

$$ A_l = -\frac{\rho_s - \rho'}{\rho_s} \frac{R^{l+2}}{\rho} \left( \frac{\zeta}{R} + \frac{2\dot{R}}{R} \right), $$

$$ B_l = -\frac{R^{l+2}}{\rho} \left( \frac{\zeta}{R} + \frac{2\dot{R}}{R} \right). $$

Employing relation for the pressure and the dependence between growth rate and chemical potential difference, we obtain for the undisturbed growth of the solid phase

$$ \rho_{ct} \left( R \dot{R} + \frac{3}{2} \dot{R}^2 \right) + \frac{\rho'}{\rho} \dot{R} = \frac{\rho' - \rho}{\rho} \left( \Delta P - \frac{\rho}{\rho' - \rho} \dot{R} \right). $$

The growth equation looks like the motion of a particle with the effective mass $M(R) = 4\pi\rho_{ct} R^2$, drag force $4\pi R^2 \rho K^{-1} R$, and potential energy $U(R) = 4\pi\alpha R - ((\rho' - \rho)/\rho) \Delta P (4\pi R^2/3)$.

The small deformations for the spherical surface of the solid phase are described by the relation

$$ \rho_{ct} \left( R \dot{R} + \frac{3}{2} \dot{R}^2 \right) + \frac{\rho'}{\rho} \dot{R} \dot{\zeta} + \frac{1}{\rho} \left( \frac{\alpha}{\rho} \frac{(l - 1)(l + 2)}{R^2} - \frac{\rho_{ct}}{l + 1} \dot{R} \right) \zeta = 0. \tag{1} $$

For $\dot{R} = \ddot{R} = 0$ and in the lack of energy dissipation, the equation describes the spectrum of crystallization waves on the spherical interface

$$ \omega_{cl}^2 = \frac{\alpha}{\rho_{ct} R^2} (l - 1)(l + 1)(l + 2). $$

The asymptotic behavior of function $\zeta_l(t)$ at large $t$ is given by

$$ \zeta_l(t) \sim \exp \left( \int_0^t dt' \left[ -\lambda_l(t') + \sqrt{\lambda_l^2(t') + \lambda_l(t') - \nu_l(t')} \right] \right). $$
where

$$\lambda_l(t) = \frac{3}{2} \frac{\dot{R}}{R} + \frac{1}{2} \frac{\rho'}{\rho_{ef}} \frac{l + 1}{KR},$$

$$\nu_l(t) = \omega_{0l}^2 - (l-1) \frac{\dot{R}}{R}.$$  

For the fixed time instant, the proper frequencies are determined by equation (1). The onset of instability at the fixed time occurs if the imaginary part of the frequency becomes positive. To estimate, we consider the perturbation as $\zeta_l \sim \exp(-i\omega t)$. In the dimensionless units

$$\Omega = \frac{\omega}{\omega_{0l}}, \quad \xi = \frac{1}{2\omega_{0l}} \left(3 \frac{\dot{R}}{R} + \frac{\rho'}{\rho_{ef}} \frac{l + 1}{KR} \right), \quad \eta = \frac{l - 1}{\omega_{0l}} \frac{\dot{R}}{R},$$

the proper frequencies are determined from

$$\Omega_{1,2} = -i\xi \pm \sqrt{1 - \eta - \xi^2}.$$  

The condition of appearing the instability is $\text{Im}(\Omega) > 0$. The result is given in Fig. 2. Provided that $1 - \eta - \xi^2 > 0$, the frequencies have the real part but the instability appears for the negative value of parameter $\xi$. If the radicand is negative, the both frequencies are imaginary. In this case the left half-plane of the phase diagram corresponds also to instability. The appearance of the instability takes place in the right half-plane for $\eta > 1$.

In Fig. 3 a simulation is shown for the behavior of spherical harmonics at the initial stage of the abnormal crystal growth. The variation of the crystal radius is described approximately by expression $\dot{R} = R_0 \sin 2\pi f_0 t$. The following values $R_0 = 0.1$ cm, $K = 0.1$ s/cm, $f_0 = 2.5$ kHz of experimental parameters at 0.5 K and overpressure 6 mbar are taken to calculate. The other quantities are the tabular data. The deformation of unit amplitude is put for the initial values. The velocity is varied within (-100, 100). In the figure the solutions are given with zero initial velocity. Note that the variation of the initial velocity in the range analyzed does not lead to the qualitative change in the behavior of the curves with the exception of first move at which fast relaxation occurs. Next in all cases the damping of the harmonics, checked to $l = 200$, is observed. The increase of the degree of the harmonic, as it follows from the simulation, results in the faster relaxation of the deformation. Thus, the numerical simulation agrees with the previous conclusion about the crystal interface stability at the initial growth stage.

**Summary.** Non-uniformity of the crystal growth results in new qualitative hydrodynamic instability of the liquid-solid interface. At the plane interface the development of instability starts only for the accelerated growth of the solid phase when the magnitude of the interfacial acceleration exceeds the critical one determined by the acceleration of gravity.

For free growth of the solid phase when its shape is almost spherical, the conditions of appearing the instability are determined both by the interfacial acceleration and by the growth rate. The experimental situation in which one may expect an observation of such instability is realized with the abnormal helium crystal growth when the interfacial mobility increases drastically by several orders of the magnitude [13] and the interfacial velocity
and acceleration reach the huge magnitudes as is noted in the Introduction. For lowest mode \( l = 2 \), the normalized order-of-magnitude parameters are about \( \xi \sim 10^2 \) and \( \eta \sim -10^4 \). However, during the first half-cycle of the oscillating growth, as is seen from Fig. 2, the trajectory of the crystal growth lies in the fourth quadrant in which the instability considered here does not appear. Thus the involvement of the non-uniform interfacial motion alone may fail to explain the liquid-solid interface instability observed. Note that none of the hydrodynamic instabilities known so far can be responsible for the instability of the crystal shape observed at the abnormal crystal growth 6.

Acknowledgments. The work is supported by the RFBR Grant No. 05-02-16806.

* burmi@kurm.polyn.kiae.su

[1] M. Yu. Kagan, Zh. Eksp. Teor. Fiz. 90, 498 (1986) [Sov. Phys. JETP 63, 288 (1986)].
[2] P. Nozieres, M. Uwaha, J. de Physique 47, 263 (1986).
[3] L. A. Maksimov, V. L. Tsymbalenko, Zh. Eksp. Teor. Fiz. 122, 530 (2002) [JETP 95, 455 (2002)].
[4] V. L. Tsymbalenko, Fiz. Nizk. Temp. 21, 162 (1995) [Low Temp. Phys. 21, 120 (1995)].
[5] V. L. Tsymbalenko, J. Low Temp. Phys. 121, 53 (2000).
[6] V. L. Tsymbalenko, Zh. Eksp. Teor. Fiz. 126, 1391 (2004) [JETP 99, 1214 (2004)].
[7] G. I. Taylor, Proc. Roy. Soc. (London) A201, 192 (1950).
[8] G. Birkhoff, Hydrodynamics, (Princeton Press, 1960).
[9] L. D. Landau, E. M. Lifshits, Fluid Mechanics, (Pergamon Press, 1987), §140.
[10] A. F. Andreev, V. G. Knizhnik, Zh. Eksp. Teor. Fiz. 83, 416 (1982) [Sov. Phys. JETP 56, 226 (1982)].
[11] C. D. Demaria, J. W. Lewellen, A. J. Dahm, J. Low Temp. Phys. 89, 385 (1992).
[12] S. Balibar, H. Alles, A. Ya. Parshin, Rev. Mod. Phys. 77, 317 (2005).
[13] V. L. Tsymbalenko, J. Low Temp. Phys. 138, 795 (2005).