Rayleigh-Taylor instability of crystallization waves at the superfluid-solid ⁴He interface

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At the superfluid-solid ⁴He interface there exist crystallization waves having much in common with gravitational-capillary waves at the interface between two normal fluids. The Rayleigh-Taylor instability is an instability of the interface which can be realized when the lighter fluid is propelling the heavier one. We investigate here the analogs of the Rayleigh-Taylor instability for the superfluid-solid ⁴He interface. In the case of a uniformly accelerated interface the instability occurs only for a growing solid phase when the magnitude of the acceleration exceeds some critical value independent of the surface stiffness. For the Richtmyer-Meshkov limiting case of an impulsively accelerated interface, the onset of instability does not depend on the sign of the interface acceleration. In both cases the effect of crystallization wave damping is the reduction in the perturbation growth rate of the Taylor unstable interface.

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I. INTRODUCTION

The Rayleigh-Taylor instability is a fingering instability of an interface between two fluids of different densities. It takes place when the heavier fluid is decelerated by the lighter fluid or, in other words, density and pressure gradients have opposite directions [1,2]. The Rayleigh-Taylor instability [3] occurs in numerous physical and technological situations, e.g., gravity-driven instability of a heavier fluid atop a lighter one and inertial confinement fusion. In essence, the Rayleigh-Taylor instability is the first step in a fluid-mixing mechanism, the step eventually leading via formation of bubbles, spikes, and curtains to the turbulent regime of fluid mixing. Along with the Kelvin-Helmholtz criterion for tangential discontinuities at the interface between two normal fluids, the Rayleigh-Taylor criterion is among the most generic principles in the complicated subject of interface instability.

For the interfaces of superfluid ⁴He, some of the hydrodynamic instabilities have been found as well. First, we mention the Faraday instability which denotes the phenomenon of the parametric excitation of standing waves on the free surface of a fluid. The flat shape of the surface becomes unstable with a periodic modulation of the acceleration of gravity. Recently [4], generation of Faraday standing waves on the free surface of ⁴He has been realized in the experimental cell subjected to sinusoidal vibration in the vertical direction. According to [5,6], it is also possible to generate a dense fog of helium droplets by driving the capillary waves on a superfluid ⁴He surface unstable with an intense ultrasonic beam from a piezoelectric transducer under the surface. There have been observed electrohydrodynamic interface instabilities due to charges trapped at the surfaces and interfaces of various condensed helium phases [7]. The shear flow between the superfluid A and B phases of 3 He can result in the Kelvin-Helmholtz interface instability [8].

Dynamics of the superfluid-solid ⁴He interface due to sufficiently fast processes of crystallization and melting resembles much that of the free surface of a fluid. In particular, as was predicted by Andreev and Parshin, the crystal in contact with its liquid phase can support wavelike processes of crystallization and melting (see review [9]). From the dynamical point of view such weakly damping crystallization waves are an immediate counterpart of the well-known gravitational-capillary waves at the vapor-liquid interfaces.

A series of mechanical and hydrodynamical instabilities has been predicted and observed for the superfluid-solid ⁴He interface. We mention the Grinfeld instability under uniaxial stress of a solid. Warping of the flat interface occurs at some threshold stress when the release of elastic energy exceeds the loss of the surface energy [9,10]. Like normal fluids, the steady flow of a superfluid in the direction tangential to the interface can result in the Kelvin-Helmholtz instability. As the flow exceeds a threshold magnitude, crystallization waves appear at the superfluid-solid ⁴He interface [11,12]. The phenomenon has qualitatively been observed as a distortion of the crystal surface in the fluid jet [13]. An analogy with generating sea waves by wind is fully appropriate here.

To date, the Rayleigh-Taylor phenomena have extensively been studied in normal fluids, but not much study has been made in superfluids or quantum solids. The classical Rayleigh-Taylor instability of the superfluid-solid ⁴He interface in the field of gravity is observed by Demaria *et al.* [14]. 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 inversion a single finger of the liquid phase ascends at the center of a cell, and the solid phase descends along the walls. On the other hand, in experiment [15] the flat shape of the interface remained stable for a ⁴He crystal grown at the needle point with its lower facet under favorable condition for developing the gravity-driven instability. A difference in the observations can be associated with the following reasons. The requirement for the interface instability, as well as the initial stage of fingering process, as is shown in [16], is sensitive to the state of a crystal facet, rough or smooth, and to the size of a facet.

Recently [17], it has been demonstrated that the Faraday instability is also inherent in the superfluid-solid ⁴He inter-

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FIG. 1. The growth of a 4 He crystal at 0.47 K and initial overpressure of 5.2 mbar. The left frame corresponds to 0.19 ms after nucleation. The right frame is taken 80 ms later when the net pressure is already close to the melting pressure. The vertical size of the frames is 2.4 mm.

face. Crystallization waves at the horizontal interface between superfluid and solid ⁴He are generated by a periodic vibration in an experimental cell in the vertical direction. In accordance with expectation [18] the amplitude of the waves excited at one-half of the driving frequency decreases for higher temperatures due to reduction in the interface growth coefficient describing dissipative properties of the interface. From general arguments the Faraday instability can be viewed as a particular case of the Taylor instability for the periodically driven interface.

On the other side, the spectrum of crystallization waves remains invariable for the steady flow of a superfluid in the direction normal to the interface [12]. At first sight this implies that the growth of a solid should not bring the superfluid-solid interface to instability. However, in the experiments on free growth of a ⁴He crystal initiated at the needle point immersed into the overpressurized liquid bulk [19,20] one can observe a destruction in the regular shape of the crystal triggered under sufficiently large overpressure exceeding about 6 mbar. Immediately after nucleation the crystal seed has a clear hexagonal prismlike shape with slight ripples. Soon afterward by 0.1-0.2 ms the shape of the crystal becomes round with a highly irregular outline. Far later by 100-150 ms, as the net overpressure vanishes and the pressure in the cell becomes phase equilibrium, the shape of the grown crystal relaxes to a regular hexagonal prism. We put two images of a crystal in Fig. 1.

For overpressures higher than 6 mbar (see Fig. 2), we discover more exotic patterns by the same time interval of 0.1-0.2 ms after the voltage pulse which triggers nucleation. The interfacial irregularities become more pronounced and acquire a mushroomlike shape. Apparently, the fluid moves into the crystal between the neighboring spikes, resulting eventually in generation of liquid bubbles inside the crystal.

In addition, in Fig. 2 we show the behavior of the pressure in the course of the crystal growth. After nucleation of a crystal seed the overpressure in the cell drops and then gradually vanishes, oscillating around zero value corresponding to the equilibrium pressure. The pressure oscillations are due, in the first turn, to the finiteness of the experimental volume and the finiteness of sound velocity. The point is that the appearance of a solid seed and its next growth are accompanied by variations in the density and the volume in which the density changes. This process results in exciting and emitting sound waves which propagate in the direction



FIG. 2. The deviation of the pressure from equilibrium vs time during crystal growth at 0.48 K and initial overpressure of 7 mbar. The left insert shows a crystal at 0.64 ms after nucleation. The time at the right insert is 16 ms. The vertical size of the frames is 2.4 mm.

to the container walls with the next backward reflection from the walls to the solid seed. The excitation and emission of sound become more and more effective as the interface rate and acceleration increase. Eventually, we obtain acoustic damping oscillations of the liquid inside the cell [21]. In some sense the damping of the pressure oscillations represents a quality factor of a liquid/solid or melting/freezing resonator.

The pressure in the cell becomes equilibrium pressure in 2 ms and the driving force vanishes. Finally at 16 ms, the crystal relaxed and acquired the regular hexagonal shape. However, we still observe the liquid bubbles embedded into the crystal bulk.

If we roughly estimate the velocity which the interface should acquire by the time of the overpressure release and formation of the irregular outline, we will find a rather high magnitude of several meters per second. The corresponding acceleration which provides such increment of velocity should be about 10^3g , g being acceleration of gravity. On the whole, large acceleration for a short time shows evidence in favor of a shock-driven character of the crystal growth. It is interesting to note that the irregular patterns observed are similar in appearance to those obtained in studies [22] of a shock-accelerated boundary between two gases of different densities. The typical time of the pattern formation was of the same order of several tenths of milliseconds.

In the present work we develop a linear theory on the Taylor instability of an arbitrarily accelerated boundary between the superfluid and solid ⁴He phases. In essence, we derive an equation which the small interfacial perturbations obey. We consider three typical cases of the interface acceleration: constant, shock, and periodic. The plane and spherical interface geometries are analyzed.

II. PLANE GEOMETRY AND INTERFACE GROWTH KINETICS

Let us assume the interface is parallel to the x-y plane, with vertical position z=L(t) which moves at the rate V $=\dot{L}(t)$. The upper half-space z > L(t) is occupied with the liquid phase and the solid phase occupies the lower one z < L(t). Below we will consider the stability of the moving interface with respect to its small perturbations $\zeta(x, y, t)$ from the flat shape. Thus, the coordinate $Z(x, y, t) = L(t) + \zeta(x, y, t)$ gives the vertical position of the perturbed interface evolving in time.

To discover the effect of nonuniform motion on the stability of the interfacial shape, we make a number of simplifying assumptions which do not affect the main point of the phenomenon. The validity and criteria of applicability for the assumptions to be made below can be found in the papers on the kinetic interface coefficients and crystallization waves [23-26].

So, in the superfluid we employ the usual two-fluid equations without dissipation. In addition, we assume that the growth rate of a solid V(t) is always small compared with the velocity of the first or second sound. This is an ordinary experimental situation. Thus we treat the hydrodynamics of the superfluid in the approximation of incompressible liquid and the constancy of the entropy density. In this case [27] the velocities \boldsymbol{v}_n and \boldsymbol{v}_s of the normal and superfluid motions can be described in terms of gradient of velocity potentials ϕ_n and ϕ_s which satisfy $\nabla^2 \phi_n = 0$ and $\nabla^2 \phi_s = 0$, respectively. Since finally we will discuss only the linearized equations in the perturbation ζ , it is convenient to consider a single Fourier mode of the perturbation $\zeta = \zeta_q(t) \exp(i\mathbf{q} \cdot \mathbf{r})$ with wave vector $\mathbf{q} = (q_x, q_y)$ parallel to the boundary. The solutions of $\nabla^2 \phi_{n,s} = 0$ can be represented as

$$\phi_s = u_s(t)z + A_s(t)\exp(i\boldsymbol{q}\cdot\boldsymbol{r} - qz),$$

$$\phi_n = u_n(t)z + A_n(t)\exp(i\boldsymbol{q}\cdot\boldsymbol{r} - qz), \qquad (1)$$

where $\mathbf{r} = (x, y)$ and the velocities $u_s(t)$ and $u_n(t)$ describe the undisturbed motion in the superfluid. The perturbation amplitudes $A_s(t)$ and $A_n(t)$ are assumed to be linear in $\zeta_q(t)$ and will be determined later from the corresponding boundary conditions at the interface. The pressure in the superfluid is a sum of pressures $P = P_n + P_s$ and

$$P_{s} = P_{s\infty} - \rho_{s} [\dot{\phi}_{s} + (\nabla \phi_{s})^{2}/2] - \rho_{s} gz,$$

$$P_{n} = P_{n\infty} - \rho_{n} [\dot{\phi}_{n} + (\nabla \phi_{n})^{2}/2] - \rho_{n} gz.$$
(2)

The index " ∞ " refers to the values taken away at infinity.

Unlike previous considerations [23–26] which are also linear in the interfacial perturbations, we have to retain the quadratic terms in the superfluid and normal velocities $\boldsymbol{v}_{s,n} = \nabla \phi_{s,n}$ on account of nonzero value $u_{s,n}(t)$ and product like $u_s A_s$ or $u_n A_n$.

The mass continuity across the boundary z=Z(x, y, t) gives

$$j_{\nu} = \rho_{n} v_{n\nu} + \rho_{s} v_{s\nu} = (\rho - \rho') \dot{Z}, \qquad (3)$$

where \dot{Z} is the velocity of the boundary, $\boldsymbol{\nu}$ is the unit vector normal to the boundary, and j_{ν} is the mass current normal to the boundary. The densities ρ_n and ρ_s are the normal and superfluid densities, $\rho = \rho_n + \rho_s$, and ρ' is the density of the solid phase. The normal components of velocities can be approximated by $v_{n\nu} \approx v_{nz}$ and $v_{s\nu} \approx v_{sz}$. We also believe that there is no motion in the solid phase, i.e., v' = 0.

To further simplifications, we suppose the normal component sticks to the interface like a viscid fluid. Also, this implies the Kapitza resistance to be infinite [24]. In addition, we disregard any excitations, e.g., phonons, in the solid. So, we put at the boundary

$$v_{n\nu} = \dot{Z}.$$
 (4)

Using Eqs. (1), (3), and (4), we can determine the unknown amplitudes in Eq. (1) and then the velocity fields v_s and v_n in the liquid. For the unperturbed motion, it is obvious that

$$u_s = -\frac{\rho' - \rho_s}{\rho_s} V \quad \text{and} \quad u_n = V.$$
(5)

The amplitudes $A_n(t)$ and $A_s(t)$ are given approximately by

$$A_{s}(t) = \frac{\rho' - \rho_{s}}{\rho_{s}} \frac{\dot{\zeta}_{q}(t)}{q} e^{qL(t)},$$
$$A_{n}(t) = -\frac{\dot{\zeta}_{q}(t)}{q} e^{qL(t)}.$$

As a result, we can also calculate pressure field (2) in the liquid. Boundary conditions (3) and (4) in combination with the obvious relations [Eq. (5)] determine unambiguously the inertial properties of the interface described in terms of the effective density [24]

$$\rho_{\rm ef} = \rho_n + (\rho' - \rho_s)^2 / \rho_s$$

in the sense that

$$\rho_s u_s^2 / 2 + \rho_n u_n^2 / 2 = \rho_{\rm ef} V^2 / 2.$$

To proceed further, we adopt the most simplifying assumptions [24] to describe the solid and its boundary. The solid is assumed to be always unstressed and all possible shearing components $\sigma_{i\neq k}$ of the stress tensor σ_{ik} are neglected. In other words, the stress tensor is isotropic, i.e., $\sigma_{ik} = -P' \delta_{ik}$, and we can define "pressure" according to P' $= -\sigma_{ii}/3$ [26]. Then, from the formal point of view, the solid can be described as a liquid under pressure equal to P'.

The next boundary condition stems from the continuity of the momentum flux density across the interface. The momentum flux density in the superfluid [27] reads $P \delta_{ik} + \rho_n v_{ni} v_{nk}$ $+ \rho_s v_{si} v_{sk}$. Then, we take $\sigma_{ik} v_k = -P' v_i$ into account, assume the small curvature of the interface z = Z(x, y, t) = L(t) $+ \zeta(x, y, t)$, and use a frame that refers to the boundary

$$P + \rho_n (\boldsymbol{v}_n - \boldsymbol{Z}\boldsymbol{\nu})^2 + \rho_s (\boldsymbol{v}_s - \boldsymbol{Z}\boldsymbol{\nu})^2 - (P' + \rho' \boldsymbol{Z}^2)$$

= $\gamma_{ik} \partial \boldsymbol{Z}^2 / \partial r_i \partial r_k = \gamma_{ik} \partial \boldsymbol{\zeta}^2 / \partial r_i \partial r_k.$ (6)

Neglecting the quadratic terms in velocities gives the usual Laplace condition of mechanical equilibrium across the interface [24]. Here $\gamma_{ik}(\theta, \varphi) = \alpha \delta_{ik} + \partial \alpha^2 / \partial \varphi_i \partial \varphi_k$ is the surface stiffness tensor [9,10,24] expressed in terms of surface tension $\alpha = \alpha(\theta, \varphi)$ depending on the angles between the crystalline orientation and the normal to the surface.

Let us turn now to the last boundary condition. It is a reasonable assumption that any motion of the interface accompanied also by the melting and growth of a solid will dissipate a certain amount of energy. Thus a finite velocity of the interface should produce some imbalance in the chemical potential difference $\mu - \mu'$ between the liquid and solid. The routine in various theories of the interfacial dynamics is an introduction of the so-called growth coefficient *K* which relates the interface growth rate with the difference in chemical potentials across the interface [23,24]. Because of $u_n(t) \neq 0$ and $u_s(t) \neq 0$ we again have to take into account the squares of velocities which are always omitted in the linear perturbation theory of the interface being initially at rest. So, at the boundary we employ an effective relation

$$\dot{Z} = K \left[\mu + \frac{(\boldsymbol{v}_s - \dot{Z}\boldsymbol{\nu})^2}{2} - \left(\mu' + \frac{\dot{Z}^2}{2} \right) \right],$$
 (7)

where μ and μ' are the chemical potentials of the liquid and solid per unit mass. In a wide sense the growth coefficient here is a certain combination of all Onsager coefficients and the kinetic coefficients describing the near-surface dissipative processes. In general, the growth coefficient *K* can depend on the temperature as well as on the wave vector *q*. Usually, in the ballistic regime, when the mean free path *l* of excitations is large, the growth coefficient is independent of wave vector. In the opposite hydrodynamic limit $ql \ll 1$ the growth coefficient may depend on the wave vector approximately as $1/K \sim ql$ [24].

Lastly, we need an expression for the chemical potential difference. As usual, the reference point is the melting pressure P_c at which the chemical potentials μ and μ' coincide and the liquid-solid transition takes place. We take the necessary formulas for the superfluid from Ref. [27]. After expanding chemical potentials in the vicinity of the melting pressure, we obtain

$$\mu - \mu' = \sigma(T - T_{\infty}) + \frac{P - P_c}{\rho} - \frac{\rho_n (\boldsymbol{v}_n - \boldsymbol{v}_s)^2}{2} - \frac{P' - P_c}{\rho'},$$
$$T - T_{\infty} = \frac{\rho_n}{\sigma \rho} \left(\frac{P_n - P_{n\infty}}{\rho_n} - \frac{P_s - P_{s\infty}}{\rho_s} - \frac{(\boldsymbol{v}_n - \boldsymbol{v}_s)^2}{2} \right),$$

where σ is the entropy and the quantities with index ∞ stand for the magnitudes taken far from the interface.

Now we are in position to find the equations which the interface dynamics obeys. Knowing velocity potentials ϕ_n and ϕ_s expressed via $u_{n,s}(t)$ and perturbation $\zeta_q(t)$, we can calculate the normal and superfluid velocities, pressure, and chemical potential difference. Next, we insert the quantities calculated at the interface into boundary conditions (6) and (7) and eliminate the pressure P'. As a result of some algebraic formula manipulation linear in ζ_q , we obtain an equation consisting of the ζ_q -independent component and the other one linear in ζ_q . The first component gives an equation

$$V\frac{\rho'}{K} = \frac{\rho' - \rho}{\rho} \left[\Delta P - \rho gL \right] + \rho_{\rm ef} \left(\dot{V}L + \frac{V^2}{2} \right)$$

which describes the undisturbed motion of the flat interface and relates overpressure $\Delta P(t) = P_{\infty}(t) - P_c$ to $V(t) = \dot{L}(t)$ in a complicated manner in order to support the necessary behavior of the growth rate. This equation does not have much interest for us.

The other equation obtained is the most significant one. It represents the equation for the linear dynamics of the interface perturbation $\zeta = \zeta_q(t) \exp(i\mathbf{q} \cdot \mathbf{r})$ when the interface is subjected to an arbitrary driving acceleration $\dot{V}(t)$,

$$\rho_{\rm ef} \frac{\zeta_q}{q} + \frac{\rho'}{K} \dot{\zeta}_q + \left[\gamma_{ik} q_i q_k + (\rho' - \rho)g - \rho_{\rm ef} \dot{V}(t)\right] \zeta_q = 0.$$
(8)

The new aspect of the equation derived is an additional term with the interface acceleration $\dot{V}(t)$. As is expected, the uniform motion of the interface at V(t)=const does not influence the character of small interfacial oscillations. For $\dot{V}(t)$ =0, Eq. (8) amounts to the known relation determining the spectrum of crystallization waves when the meltingcrystallization processes are balanced and the interface position in average is invariable, i.e., L(t)=const [9,24,25].

Undoubtedly, more realistic and complicated models of the superfluid-crystal ⁴He interface will improve the magnitudes of the effective interface density, effective growth coefficient, and surface stiffness. However, we believe that the structure of Eq. (8) is generic and holds.

III. INTERFACE INSTABILITIES

Equation (8) can have unstable solutions depending strongly on the acceleration history of the interface. First, let us consider the stability of the plane interface with respect to small perturbations $\zeta_q \sim \exp[\lambda(q)t]$ for the uniformly accelerated growth of a crystal. The root with Re $\lambda(q) > 0$ means the interface instability, i.e., initially small-amplitude perturbations of wavelength $2\pi/q$ will grow exponentially in time. For the acceleration exceeding the threshold $V_c = g(\rho')$ $-\rho)/\rho_{\rm ef}$, the interfacial perturbation will increase for the wave vectors satisfying $\gamma q_c^2 < \rho_{\rm ef} \dot{V} - (\rho' - \rho)g$. For brevity, we put $\gamma_{ik}q_iq_k = \gamma q^2$ where $\gamma = \gamma_{ik}n_in_k$ and n = q/q is unit vector in the direction of perturbation. Note that the threshold acceleration 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 V is directed to the superfluid. Thus, the Taylor instability due to nonuniform growth of the plane interface appears only for the accelerated growth of a solid.

The surface stiffness stabilizes the region of long wave perturbations and establishes the most unstable wavelength having the fastest exponential growth. The value q_0 corresponding to the maximum magnitude Re $\lambda(q)$ gives the shortest time scale for the development of the instability which will be characterized by the wavelength $2\pi/q_0$. The value q_0 can be found from the equation

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$$q_0^2 = \frac{q_c^2}{3} - \frac{\sqrt{2}}{3} \frac{\rho'}{\sqrt{\gamma \rho_{\rm ef}}} \frac{q_0^{3/2}}{K},$$

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

$$q_0 = \frac{q_c}{\sqrt{3}} \left(1 - \frac{\rho'}{(6\sqrt{3}\gamma\rho_{\rm ef}K^2q_c)^{1/2}} \right),$$
$$\lambda_0 = \left(\frac{2\gamma}{3\sqrt{3}} \frac{q_c^3}{\rho_{\rm ef}} \right)^{1/2} - \frac{q_c}{2\sqrt{3}} \frac{\rho'}{\rho_{\rm ef}K}.$$

The values q_0 and λ_0 depend on γ , thus implying anisotropic and complicated possible surface patterns.

In the opposite limit $K^4 \dot{V} \ll \rho'^4 / \gamma \rho_{\rm ef}^3$ one has roughly

$$q_0 = \left(\frac{\gamma \rho_{\rm ef}}{\rho'^2} K^2 q_c^4\right)^{1/3},$$
$$\lambda_0 = \frac{K}{\rho'} \gamma q_c^2 = \frac{K}{\rho'} [\rho_{\rm ef} \dot{V} - (\rho' - \rho)g].$$

It is interesting that, though the spatial scale q_0^{-1} is sensitive to the surface stiffness and its anisotropy, the time of developing the instability becomes independent of the surface stiffness and its anisotropy.

On the whole, the values q_0 and λ_0 decrease as the kinetic growth coefficient reduces or dissipation with the interface enhances. From the experimental point of view this may require a crystal surface of sufficiently large sizes $d > 2\pi/q_0$ and a large time to support the accelerated growth $t > 1/\lambda_0$ in order to realize an interfacial instability with a uniformly accelerated growth.

In Ref. [18] it has been shown that periodic modulation of the gravitational constant as $g(t)=g(1+2\tilde{\epsilon}\cos 2\omega t)$ can result in parametric excitation of crystallization waves at the stationary flat interface corresponding to V(t)=0 in our case. Admitting some analogy between gravity and noninertial frame, one can expect a possibility of exciting crystallization waves with a periodic driving, e.g., $\dot{V}(t)=G\cos 2\omega t$, even with the lack of gravity. To demonstrate this, let us rewrite Eq. (8) in the form

$$\ddot{\zeta}_q + \Gamma_q \dot{\zeta}_q + \omega_0^2(q) [1 - \dot{V}(t)/\tilde{g}] \zeta_q = 0, \qquad (9)$$

where we have introduced the damping coefficient Γ_q , the frequency $\omega_0(q)$ of crystallization waves in the lack of damping, and the scaled accelerating amplitude \tilde{g} ,

$$\omega_0^2(q) = [\gamma_{ik}q_iq_k + (\rho' - \rho)g]q/\rho_{\text{ef}}, \quad \Gamma_q = \frac{\rho'}{\rho_{\text{ef}}}\frac{q}{K},$$

$$\widetilde{g} = g \frac{(\rho' - \rho)}{\rho_{\rm ef}} \left(1 + \frac{\gamma_{ik} q_i q_k}{(\rho' - \rho)g} \right).$$

Then, if a periodic process of melting and crystallization is realized in experiment so that the interface could oscillate around some average position at frequency 2ω and amplitude $G/(2\omega)^2$, Eq. (9) with label $2\epsilon = -G/\tilde{g}$ transforms to a Mathieu-type equation,

$$\ddot{\zeta}_q + \Gamma_q \dot{\zeta}_q + \omega_0^2(q)(1 + 2\epsilon \cos 2\omega t)\zeta_q = 0,$$

which is identical to that analyzed in [18]. The predictions which follow are well known and we refer the readers to papers [17,18] for details. Note only that in the free crystal growth experiments [15,20] the pressure in the cell drops drastically down after nucleating a solid seed with the subsequent transition to the damped oscillations around the melting pressure.

Crystallization waves at the superfluid-solid ⁴He interface can also be generated with the Richtmyer-Meshkov mechanism when the interface is subjected to an impulsive acceleration, i.e., $\dot{V}(t) \sim V(0) \,\delta(t)$. In ordinary fluids and gases, for this purpose, the passage of a shock wave across the interface is commonly used. Unlike the Taylor case of constant acceleration when the perturbation amplitude in the linear regime grows exponentially in time, the initial stage of interface instability in the Richtmyer-Meshkov case of shock acceleration [28] is characterized by a linear growth of the perturbation amplitude in time. The Richtmyer-Meshkov instability is independent of the direction of acceleration in contrast to the Taylor one. The late time stages of both instabilities may demonstrate a formal resemblance, showing bubble and spike morphology.

According to Eq. (8), the growth rate of the Richtmyer-Meskov unstable interface in the linear regime can approximately be described by

$$\begin{aligned} \zeta_q(x,t) &= \frac{qV(0)}{\Gamma_q} (1 - e^{-\Gamma_q t}) \zeta_q(x,0) \\ &= \frac{\rho_{\rm ef}}{\rho'} K V(0) (1 - e^{-\Gamma_q t}) \zeta_q(x,0), \end{aligned}$$
(10)

where $\zeta_a(x,0) = \zeta_a(0) \cos qx$ is the initial perturbation amplitude at the interface immediately after the impulsive acceleration and V(0) is an increment in the interface velocity due to acceleration of the boundary. At the linear stage the amplitude of crests and troughs is approximately the same. The shape of the crests and troughs is similar and the interface remains approximately sinusoidal. Staying in the linear regime, we see from Eq. (10) that the finite damping of crystallization waves is a stabilizing factor and can limit the growth of the perturbation amplitude. It is worthwhile to note that the total amplification factor is proportional to the interface growth coefficient K and proves to be independent of the wavelength $2\pi/q$ of an initially sinusoidal perturbation. On the contrary, the time for ceasing the growth of perturbation amplitude is scale dependent. The larger the wavelength is, the longer the decay time is. A special feature of crystallization waves in ⁴He is that the growth coefficient K, and thus Γ_q , is strongly temperature dependent. In ⁴He the Richtmyer-Meshkov instability can be studied with a sound wave pulse hitting the interface in the normal direction. Recently [29], it has experimentally been demonstrated that the superfluid-solid ⁴He interface can be set in motion with a sound wave which transmits through the interface, giving rise to the processes of crystallization and melting.

IV. SPHERICAL GEOMETRY

In experiment [20] a solid nucleates at the needle point and then grows free. A ratio of the crystal sizes in the different directions for the crystals grown is not drastic. The ratio of the maximum to minimum size does not exceed 2–3 (see also Figs. 1 and 2). Here we consider the stability of the spherical shape of a growing solid. For simplicity, we assume the surface tension α is isotropic. In addition, we neglect the acceleration of gravity and, as above, treat the equations linearized in the interface perturbation.

The equation specifying the interface motion is taken as $r=R_s(t,\Omega)=R(t)+\zeta(t,\Omega)$, where R(t) is the radius of the unperturbed spherical interface and $\zeta(t,\Omega)=\sum_l \zeta_l(t)Y_l(\Omega)$ is an interface perturbation expanded in the spherical harmonics of degree l=0,1,2,... We look for velocity potentials of the normal $\boldsymbol{v}_n=\nabla\phi_n$ and superfluid $\boldsymbol{v}_s=\nabla\phi_s$ motions of the forms

$$\phi_s = -u_s(t)R^2/r + A_l(t)Y_{l'}/r^{l+1},$$

$$\phi_n = -u_n(t)R^2/r + B_l(t)Y_{l'}/r^{l+1}.$$

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

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

and coefficients $A_l(t)$ and $B_l(t)$ describing the perturbed motion of the interface,

$$A_{l} = \frac{\rho' - \rho_{s}}{\rho_{s}} \frac{R^{l+2}}{l+1} \left(\dot{\zeta}_{l} + \frac{2\dot{R}}{R} \zeta_{l} \right),$$
$$B_{l} = -\frac{R^{l+2}}{l+1} \left(\dot{\zeta}_{l} + \frac{2\dot{R}}{R} \zeta_{l} \right).$$

Employing the same boundary relation for the pressures in the phases and the same dependence between growth rate and chemical potential difference, we obtain for the undisturbed growth of the solid phase after some algebra,

$$\rho_{\rm ef}\left(R\ddot{R} + \frac{3}{2}\dot{R}^2\right) + \frac{\rho'}{K}\dot{R} = \frac{\rho' - \rho}{\rho}\left(\Delta P - \frac{\rho}{\rho' - \rho}\frac{2\alpha}{R}\right).$$

The growth equation looks exactly like the motion of a particle with the effective mass $M(R)=4\pi\rho_{\rm ef}R^3$, drag force $4\pi R^2 \rho' K^{-1}\dot{R}$, and potential energy $U(R)=4\pi\alpha R^2$ $-[(\rho'-\rho)/\rho]\Delta P(4\pi R^3/3), \Delta P=P_{\infty}-P_c$ being overpressure.

The small-amplitude perturbations for the spherical surface of the solid phase are described by the relation

$$\rho_{\rm ef} \left(\frac{R}{l+1} \ddot{\zeta}_l + \frac{3}{l+1} \dot{R} \dot{\zeta}_l \right) + \frac{\rho'}{K} \dot{\zeta}_l + \left(\alpha \frac{(l-1)(l+2)}{R^2} - \rho_{\rm ef} \frac{l-1}{l+1} \ddot{R} \right) \zeta_l = 0. \quad (11)$$

For large radius *R* and $l \rightarrow \infty$ so that q = l/R is fixed, Eq. (11) goes over to the result for the plane geometry. Under steady conditions $R(t) \equiv R$ the spectrum of crystallization waves on the spherical interface is determined from

$$\omega_l^2 + i\omega_l \frac{\rho'}{\rho_{\rm ef}K} \frac{l+1}{R} - \frac{\alpha}{\rho_{\rm ef}R^3} (l-1)(l+1)(l+2) = 0.$$

Equation (11) can have unstable solutions giving rise to the generation of crystallization waves at the interface. The answer whether the spherical interface will be stable or unstable depends strongly on the history of the time behavior R(t). To illustrate, we consider the Richtmyer-Meshkov situation when a solid with radius R and interface rate \dot{R} is subjected to a spherical shock acceleration $\ddot{R}=V(0)\delta(t)$. In the linear regime the perturbation amplitude can be estimated according to

$$\zeta_l(t) = \frac{V(0)(l-1)}{3\dot{R} + (l+1)\rho'/(\rho_{\rm ef}K)} [1 - e^{-(3\dot{R}/R + \rho'(l+1)/R\rho_{\rm ef}K)t}] \zeta_l(0).$$

Here $\zeta_l(0)$ is the initial perturbation amplitude of *l*th harmonic at the interface immediately after the shock acceleration and V(0) is an additional velocity acquired by the interface. The mode with l=1 is obviously not involved because this harmonic describes the displacement of a sphere as a whole. Like the case of the planar geometry, the initial growth of the interface perturbation is a linear function of time,

$$\zeta_l(t) = tV(0)(l-1)\zeta_l(0)/R.$$

The long-time behavior of the interface perturbation is strongly governed by the magnitude and the sign of the growth rate \dot{R} . In fact, provided that a solid ⁴He globe either grows at any rate or melts not so fast at the moment of shock acceleration, i.e.,

$$\dot{R} > -\frac{1}{3} \frac{\rho'(l+1)}{\rho_{\rm ef}K}$$

the growth of the perturbation amplitude saturates. For larger harmonics, the stabilizing role of the finite damping of crystallization waves increases. In the opposite regime when a ⁴He solid melts sufficiently fast, the linear growth of the interface amplitude in time will cross over into an exponential increment of the perturbation amplitude.

The effects in the spherical geometry can be studied by focusing a high-intensity sound wave in the middle of an experimental cell. The experiments [30] on nucleation of solid ⁴He with two hemispherical piezoelectric transducers glued together to make a spherical geometry have shown that it is possible to achieve very high pressure amplitudes, more than 100 bar in the bulk liquid ⁴He.

V. SUMMARY

We have investigated here the analog of the Rayleigh-Taylor and Richtmyer-Meshkov instabilities of crystallization waves at the accelerated superfluid-solid ⁴He interface. Our analysis, made within the linear theory in perturbation, shows that the Rayleigh-Taylor and Richtmyer-Meshkov instabilities can occur, as well as the parametric Faraday instability [18], which we have treated as a periodically driven version of the Taylor instability. The plane and spherical interfaces are considered, and the first-order linearized equations are found for the perturbation amplitudes.

Unfortunately, it is difficult to make a well-founded conclusion in favor of destructing the crystal faceting as a result of the impulsively accelerated interface. For a quantitative analysis, it is necessary to have the images of crystal evolution taken successively in time. However, such aspects as the Taylor-type instability cannot be rejected for the highly accelerated superfluid-solid ⁴He boundaries.

Regarding the well-faceted and atomically smooth crystal surfaces which may have an infinitely large surface stiffness, we can make the following remarks. The large value of surface stiffness γ is a factor, first, for nonzero wave vectors q, which prevents the development of the Taylor instability. The most distinctive feature of the smooth crystal surface from the rough one is the existence of a nonanalytic cusplike behavior in the angle dependence for the surface tension, e.g., [9] and [10]. The presence of a singularity leads to qualitative distinctions in the development of instabilities [16]. First and foremost, the threshold magnitude and the conditions determining the development of the Taylor instability prove to be dependent on the initial amplitude of the interfacial perturbation. The smaller the initial perturbation amplitude, the larger the necessary threshold magnitude of acceleration,

approximately as $1/\zeta_q(0)$. We note here that the onset of the Taylor instability is favored at the vicinal surfaces whose orientations are tilted by a small angle with respect to the high-symmetry faceted ones. The larger the slope of the vicinal plane, the smaller the initial amplitude of interfacial perturbations is required for the development of the instability at the same magnitudes of acceleration.

The constant acceleration of an interface works like an effective gravity. Hence, the physical picture for the Taylor case of constant acceleration is analogous to the gravity-driven fingering of a crystal atop a fluid and can qualitatively be interpreted in terms of an effective amplitude-dependent stiffness $\gamma \sim 1/q\zeta_q$ [16]. For sufficiently large perturbation amplitudes ζ_q , the difference between the cases of smooth and rough crystal surfaces disappears.

In the case of impulsively accelerated smooth interface, the very initial stage of instability will be similar to that of a rough surface with the perturbation amplitude increasing linearly in time. Insensitivity to the surface state results from the fact that the inertial properties of the interface, associated with its effective density, are mainly responsible for a linear response on instant loading. However, the specific time when the linear time growth of perturbations breaks down becomes amplitude dependent for the smooth surfaces and shorter as compared to the rough surfaces.

Experimental and theoretical study of these effects can be useful for clarifying physical aspects of the crystal ⁴He growth under high drives and fast dynamics.

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