

## Faraday Instability of Crystallization Waves at the $^4\text{He}$ Solid-Liquid Interface

Wim van Saarloos

*Institute Lorentz, Leiden University, P.O. Box 9506, 2300 RA Leiden, The Netherlands*

John D. Weeks

*Institute for Physical Science and Technology and Department of Chemistry,  
University of Maryland, College Park, Maryland 20742-2431*

(Received 23 September 1994)

Crystallization waves exist at the  $^4\text{He}$  solid-liquid interface at low temperatures that are analogous to capillary waves at a liquid-air interface. Standing capillary waves can also be excited parametrically leading to the so-called Faraday instability. We analyze here the analog of this instability for crystallization waves. The threshold for the instability is independent of the surface stiffness at low temperatures, and therefore also independent of the crystalline anisotropy. The symmetry of surface patterns above the instability threshold, on the other hand, is argued to be sensitive to crystalline anisotropy.

PACS numbers: 67.80.-s, 64.70.Dv, 68.35.Md

In 1978, Andreev and Parshin [1] made the interesting prediction that at low temperatures the interface of a  $^4\text{He}$  crystal in contact with the melt would support weakly damped crystallization waves. These are the immediate analogs of the well-known capillary waves at the liquid-vapor interface. The existence of these waves is a result of the frictionless mass transport through the superfluid at low temperatures [1–3]. While the existence of crystallization waves was verified shortly thereafter by Keshishev, Parshin, and Babkin [4], it is only in recent years [5–7] that they have been studied systematically as a tool to probe the anisotropy of the surface stiffness of the interface as a function of crystallographic orientation. An interesting difference between crystallization waves and capillary waves is that in the latter the damping results from the dissipation in the bulk of the fluid due to viscosity, while in the low temperature regime the damping of crystallization waves arises from the kinetic growth coefficient [1–3,8–10] at the interface. This coefficient is strongly temperature dependent; as a result, the damping of crystallization waves can simply be tuned by changing the temperature. Another important difference is that the crystal interface is anisotropic, as a result of which crystallization waves are governed by an anisotropic surface stiffness rather than the surface tension.

In all the experiments done so far, the crystallization waves are excited at one side of the interface of the He crystal, so that the waves propagate *along* the interface. By measuring the wavelength and the decay of the amplitude along the interface as a function of frequency, the dispersion relation can be extracted, and the surface stiffness and damping coefficient can be obtained. However, for regular fluids, it is well known that *standing* capillary waves can also be excited by a parametric instability, the so-called Faraday instability [11–13]. It is the purpose of this article to explore the crystallization wave analog of this capillary wave instability and to point out a number

of interesting features that appear to make it worthwhile to carry out such an experiment.

The Faraday instability leading to standing surface waves occurs when a fluid is oscillated uniformly in the direction perpendicular to the interface. Essentially this amounts to a periodic modulation of the gravitational acceleration. Since the instability is parametric, the frequency  $\omega$  of the capillary mode that first goes unstable upon increasing the driving amplitude corresponds to half the driving frequency  $2\omega$ . Parametric surface waves have been studied by several groups in the last few years as an interesting example of a nonequilibrium pattern forming system [13] that can exhibit low-dimensional nonlinear dynamics as well as interesting spatiotemporal behavior: transitions between ordered and disordered or chaotic patterns, secondary instabilities, and unexpected averaging behavior [12–20]. Most of these experiments have been done on fluids with a relatively small viscosity, so that the damping of the waves is small. In the small damping limit, which is the case that has been analyzed in most detail theoretically [13,21], theory predicts that square standing wave patterns should occur just above the threshold for instability; experimentally not only square [14,19,20], but also hexagonal [15] and one-dimensional striped [18], patterns have been observed close to threshold. It appears that square patterns are favored for small viscosity fluids, while other symmetry patterns are favored at higher viscosities, but this issue is not very well understood.

Experiments on parametrically excited crystallization waves offer an interesting route to help clarify this issue: as mentioned earlier, over much of the experimentally accessible temperature range the damping of crystallization waves is determined by the kinetic coefficient at the interface. The strong temperature dependence of this coefficient allows one to vary the damping without changing the other parameters of the system. In effect, one can

study the Faraday instability with a fluid with continuously tunable viscosity. In addition, the anisotropy of the surface stiffness of vicinal surfaces gives rise to surprising new effects, whose study may yield new insight into the properties of the  $^4\text{He}$  crystal-liquid interface.

For our discussion of the parametric crystallization waves, we follow Andreev and Parshin [1] in our derivation of the linearized interface equations. In the low temperature regime, the normal fluid density can be neglected; furthermore, at temperatures low enough that the growth resistance of the interface is dominated by the reflection of ballistic phonons, the growth kinetics takes the form of a local expression for the normal interface growth velocity  $v_n$  [1,8,9]

$$v_n = K(\mu_s - \mu_c). \quad (1)$$

Here  $\mu_s - \mu_c$  is the difference in the chemical potential between the superfluid and the solid. For reflection of ballistic phonons, one expects  $K$  to vary with temperature as  $T^{-4}$ , and according to the data [6,10,22] this local growth regime extends up to temperatures of several hundred millikelvin.

Let  $z = \zeta$  denote the vertical position of the perturbed interface relative to the equilibrium position  $z = 0$ ; to linear order, we then have  $v_n = \dot{\zeta}$  in (1). Since we will only discuss the linearized equations, it is convenient to consider a single mode  $\zeta = \zeta_k(t)e^{ikx}$  with  $k > 0$ . In the superfluid, we have potential flow with  $\vec{v}_s = \vec{\nabla}\psi$ . To relate  $\psi$  to the interface perturbation  $\zeta$ , we note that the frequency of crystallization waves is low in comparison with (second) sound waves. We can therefore take the superfluid incompressible,  $\vec{\nabla} \cdot \vec{v}_s = \nabla^2\psi = 0$ . In terms of Fourier modes satisfying this condition, we then have  $\psi = \psi_k(t)e^{ikx - kz}$ . For the  $z$  component of the superfluid velocity at the interface, this gives  $v_{sz} = -k\psi_k(t)e^{ikx}$ . Conservation of mass at the interface requires that  $\rho_s v_{sz} = -(\rho_c - \rho_s)\dot{\zeta}$ , where  $\rho_c$  and  $\rho_s$  are the mass densities of the crystal and the superfluid. A combination of these two results then gives

$$\psi_k(t) = \frac{\Delta\rho}{k\rho_s} \dot{\zeta}_k(t), \quad (2)$$

where  $\Delta\rho = \rho_c - \rho_s$  is the density difference between the crystal and the superfluid.

To evaluate the right hand side of (1), we note that since temperature variations can be neglected [8], variations in the chemical potential can be expressed in terms of the pressure variations at the interface,

$$\delta\mu_s - \delta\mu_c = m \left( \frac{\delta P_s}{\rho_s} - \frac{\delta P_c}{\rho_c} \right), \quad (3)$$

where  $m$  is the mass of a  $^4\text{He}$  atom. At a curved interface, there is a pressure difference that is proportional to the curvature; for the isotropic fluid-air interface, the proportionality constant is the surface tension. For the anisotropic but rough solid-liquid interface the total surface energy depends both on the surface area and on the surface orientation, and the appropriate generalization

for our case is [23]

$$\delta P_s - \delta P_c = \gamma(\phi, \theta) \frac{\partial^2 \zeta}{\partial x^2} = -\gamma(\phi, \theta) k^2 \zeta_k(t) e^{ikx}. \quad (4)$$

Here  $\gamma$  is the surface stiffness, which depends on the angles  $\phi$  and  $\theta$  between the direction of propagation of the wave and the crystalline orientation. In the Faraday experiment, the cell is oscillated in the  $z$  direction; this amounts to a modulation of the gravitational constant,  $g(t) = g(1 + \bar{\varepsilon} \cos 2\omega t)$ , where  $\bar{\varepsilon}$  is the amplitude of the modulation. For pressure variations in the superfluid at the interface, we can in the regime of interest use the Bernoulli relation to give  $\delta P_s = -\rho_s \dot{\psi} - \rho_s g(t) \zeta$ . With (2), this gives

$$\delta P_s = -\frac{\Delta\rho}{k} \ddot{\zeta}_k(t) e^{ikx} - \rho_s g(1 + \bar{\varepsilon} \cos 2\omega t) \zeta_k(t) e^{ikx}. \quad (5)$$

Using (3)–(5) for the right hand side in (1), we finally arrive at the following equation for the amplitude  $\zeta_k$

$$\ddot{\zeta}_k + \Gamma_k \dot{\zeta}_k + \omega_0^2(k) [1 + 2\varepsilon \cos 2\omega t] \zeta_k = 0, \quad (6)$$

where we have introduced the damping coefficient  $\Gamma_k$ , the frequency  $\omega_0(k)$  of crystallization waves in the absence of damping, and the scaled driving amplitude  $\varepsilon$ ,

$$\omega_0^2(k) = \frac{k\rho_s g}{\Delta\rho} \left( 1 + \frac{\gamma(\phi, \theta) k^2}{g\Delta\rho} \right), \quad \Gamma_k = \frac{\rho_c \rho_s k}{mK(\Delta\rho)^2},$$

$$\varepsilon = \frac{1}{2} \bar{\varepsilon} \left( 1 + \frac{\gamma(\phi, \theta) k^2}{g\Delta\rho} \right)^{-1}. \quad (7)$$

For a single  $k$  mode, this equation is the prototype equation describing parametric resonance, the Mathieu equation. The predictions following from this equation are well known, and they can be expressed analytically in the small damping limit  $\Gamma_k/\omega_0(k) \ll 1$ . When  $\varepsilon$  is increased, the first instability occurs at [24]

$$\varepsilon_c = \Gamma_k/\omega_0(k) (\ll 1) \quad \text{with} \quad \omega_0(k) = \omega. \quad (8)$$

Note that the frequency of the waves that first go unstable is half the driving frequency, as is characteristic of a parametric instability. Upon increasing  $\varepsilon$  above the threshold value  $\varepsilon_c$ , more and more modes with frequency  $\omega_0(k)$  near  $\omega$  become unstable, i.e., lead to exponentially growing solutions of this linear equation. In the experiments on the Faraday instability in fluids, one observes standing wave patterns slightly above threshold, because the growth of the unstable modes saturates due to nonlinearities. These standing wave patterns subsequently show transitions to more complicated states upon increasing the driving further beyond threshold [14–20]. For larger values of  $\Gamma_k$ , the scenario near threshold is essentially the same, but the resonance frequency and threshold values deviate from those given in (8) and have to be obtained numerically or from tabulated values. Experimentally one observes different patterns away from threshold in this regime [15,18].

We now discuss some of the salient differences and similarities that we expect between parametrically generated crystallization waves and capillary fluid waves, focusing on the ordered surface patterns that we expect near threshold.

(i) *Frequency and wavelength scale.* The parametric equation (7) is of the same form as the one describing the Faraday surface instability of low viscosity liquids [12,13]. Physically, the main difference is that for liquid systems, the damping term is due to the viscous friction in the bulk [25], while for crystallization waves the friction arises from the kinetic boundary condition (1) at the interface in the temperature regime we consider. There are also interesting *quantitative* differences in the parameters entering the expression for  $\omega_0(k)$ . In  $^4\text{He}$ , the density difference between the superfluid and the solid is much smaller than the analogous term for capillary waves at liquid surfaces. In the latter case  $\Delta\rho \approx \rho_l$ , the liquid density, since the density of air can be neglected. At the same time, however, the (isotropic) surface tension  $\gamma$  of typical liquid-air interfaces is a factor of 100 larger than the solid-liquid interface stiffness of the  $^4\text{He}$  interface. Thus the differences in these parameters largely cancel, and the wavelengths of capillary surface waves and crystallization waves are quite comparable. For example, the wavelength  $\lambda = 2.83$  mm that one obtains [19] by parametrically exciting a layer of silicon oil at 160 Hz, is only a factor of 2 smaller than that of crystallization waves at this frequency. We likewise expect the amplitude of parametrically excited crystallization waves to be of the same order of magnitude as those found typically in experiments on capillary fluid waves, i.e., fractions of a mm.

(ii) *Tuning the friction.* An interesting aspect of crystallization waves is that the growth coefficient  $K$ , and hence  $\Gamma_k$ , is strongly temperature dependent. Indeed, according to the data [6,7,22] at about the upper end of the temperature range where (1) is accurate,  $\Gamma_k/\omega$  is of order unity, while at lower temperatures  $\Gamma_k/\omega \ll 1$ . Contrary to the fluid case, where (6) is not accurate for ratios  $\Gamma_k/\omega$  of order unity [13,21], the Mathieu equation (6) is the proper linear equation for parametric surface waves over the whole temperature range where the kinetic expression (1) is a good approximation [2,8]. We pointed out above that the surface patterns of parametrically excited Faraday waves appear to vary with the damping ratio; it is therefore of interest to study the dependence of crystallization patterns on temperature, especially in the regime where the surface stiffness is relatively isotropic [see (iii) below].

(iii) *Anisotropy.* Let us now consider the consequences of the anisotropy in the surface stiffness  $\gamma(\phi, \theta)$ , where  $\phi$  is the angle the surface normal makes with the  $c$  axis, and  $\theta$  the angle between the projection of the  $c$  axis on the surface plane and the direction of propagation of the waves. According to the data of Rolley *et al.* [7],  $\gamma$  is essentially independent of  $\theta$  when the surface angle  $\phi \gtrsim 3^\circ$ . In this regime, we expect the nonlinear standing

wave patterns to be close to those seen in the experiments on fluids. For so-called vicinal surfaces with angles  $\phi \lesssim 3^\circ$ , however,  $\gamma$  is found to be very anisotropic; in this strongly anisotropic regime a picture of a surface that consists of many well-separated, almost parallel steps becomes appropriate. The measurements [7] show that  $\gamma$  is small for  $\theta \approx 0$  when the crystallization waves propagate in the direction perpendicular to the steps, and large for  $\theta \approx 90^\circ$  when the waves run parallel to the steps. In the anisotropic regime  $\phi \lesssim 3^\circ$ , we had intuitively expected the instability threshold to be lowest for waves with  $\theta = 0$ , i.e., waves in the direction with lowest  $\gamma$  for which surface undulations cost the lowest interfacial energy. For driving amplitudes  $\varepsilon$  just above the critical value, one would then expect the surface to consist of straight one-dimensional undulations parallel to the steps of the surface. However, the low-damping threshold condition (8) shows that this is not necessarily true. When translated back into the external driving amplitude  $\tilde{\varepsilon}$  (8), becomes

$$\tilde{\varepsilon}_c = \frac{2\rho_c \omega}{mKg\Delta\rho}, \quad \omega = \omega_0(k). \quad (9)$$

This condition is *independent* of the stiffness  $\gamma(\phi, \theta)$  and hence independent of the angle  $\theta$  of a crystallization wave relative to the crystalline orientation. This result has two important implications. First, it yields an additional, independent way to measure the growth coefficient  $K$  and to determine the regime over which (1) is an accurate approximation. Second, according to (9) all linear modes with  $\omega = \omega_0(k)$  go unstable at the same time [26]. Hence, in spite of the anisotropy, the symmetry of the standing wave pattern (e.g., rectangular or one-dimensional) just above onset of the instability, is not prescribed by the linear instability. Theoretically, the symmetry will then be determined by the nonlinear terms, which we do expect to depend sensitively on the anisotropy in  $\gamma$  and  $K$ . In particular, while recent measurements [22] show that  $K$  is indeed strongly dependent on  $\phi$ , linear crystallization wave experiments are only sensitive to the normal growth coefficient of the unperturbed flat interface and hence cannot probe the strong  $\theta$  anisotropy that one expects once deviations from planarity become important. Nonlinear wave patterns, on the other hand, will depend on this anisotropy. Experimental studies of the ordered patterns in conjunction with a nonlinear theory will therefore in principle yield new information on the surface properties of  $^4\text{He}$ . Whether the nonlinear surface pattern symmetries depend sufficiently strongly on the  $\theta$ -anisotropy of  $K$  to make this feasible, remains to be determined by a nonlinear analysis.

(iv) *Alternative excitation mechanisms.* In experiments on fluids, the modulation is usually achieved by oscillating the cell with a loudspeaker. While it is desirable to go to higher frequencies to get the wavelength  $\lambda$  much smaller than the linear cell dimension, the frequency range over which this can be done is often limited to values

around 100 Hz due to mechanical instabilities. A promising possibility, also for low temperature applications, is to oscillate the sample with a piezoelectric [27], instead of with a loudspeaker. With an amplitude modulation  $a = 1 \mu\text{m}$  that appears feasible in this way, one finds from (9) with  $\tilde{\epsilon} = 4a\omega^2/g$  that this should allow one to excite parametric waves for frequencies larger than several hundred Hz. In principle, one can also excite crystallization waves parametrically with the aid of an ac electric field  $E = \bar{E} \cos \omega t$  in the direction normal to the interface [28]. We have analyzed this situation by including the field energy terms of the form  $m\vec{p} \cdot \delta\vec{E}/\rho$  on the right hand side of Eq. (3), with the field variations  $\delta\vec{E}$  calculated to first order in the interface modulation  $\zeta_k$  and  $\vec{p}$  the polarization. The threshold field strength  $\bar{E}_{\text{th}}$  for the Faraday instability then turns out to be

$$\bar{E}_{\text{th}}^2 = \frac{4\rho_c \omega_0(k)}{mK\epsilon_0 Rk}, \quad (10)$$

with  $R = (\epsilon_c - \epsilon_s)[\rho_c \epsilon_c^2(\epsilon_s - 1) - \rho_s \epsilon_s^2(\epsilon_c - 1)]\rho_s \epsilon_s^2 \times (\epsilon_c + \epsilon_s)$ ,  $\epsilon_0$  the permittivity of free space, and  $\epsilon_c \approx 1.08$  and  $\epsilon_s \approx 1.07$  the relative dielectric constants. Because of the small difference between  $\epsilon_c$  and  $\epsilon_s$ ,  $R = \mathcal{O}(10^{-5})$ , and this gives  $\bar{E}_{\text{th}} \approx 200 \text{ kV/cm}$ . This is more than an order of magnitude larger than what appears to be experimentally feasible [29], so oscillating the sample with a piezoelectric appears to be the most promising excitation mechanism.

In conclusion, our analysis of the Faraday instability of crystallization waves shows that in the low temperature regime the threshold condition involves the growth coefficient only; in spite of the crystalline anisotropy, all surface modes go unstable simultaneously. As a result, the symmetry of ordered patterns above threshold is argued to be determined by the anisotropy through the nonlinear terms. Experimental and theoretical analysis of these effects may therefore yield new information on the surface properties of  $^4\text{He}$ .

We are grateful to R. Jochemsen for stimulating interest and for illuminating discussions, to P.C. Hohenberg and P. Nozières for useful correspondence, to S. Balibar for correspondence and for sharing unpublished data with us, and to W. van de Water for teaching W.v.S. about the Faraday instability. J.D.W. acknowledges financial support from the Netherlands Organization for Scientific Research (NWO) and the Lorentz Fund for a visit to the Institute Lorentz, which made this research possible, and from the National Science Foundation under Grant No. NSF-DMR-91-03031.

- 
- [1] A. F. Andreev and A. Ya. Parshin, Zh. Eksp. Teor. Fiz. **75**, 1511 (1978) [Sov. Phys. JETP **48**, 763 (1978)].  
 [2] A. F. Andreev and V. G. Knizhnik, Zh. Eksp. Teor. Fiz. **83**, 416 (1982) [Sov. Phys. JETP **56**, 226 (1982)].  
 [3] For a review, see A. F. Andreev, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (North-Holland, Amsterdam, 1982), Vol. VIII.

- [4] K. O. Keshishev, A. Ya. Parshin, and A. V. Babkin, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 63 (1979) [JETP Lett. **30**, 56 (1979)].  
 [5] O. A. Andreeva, K. O. Keshishev, and S. Yu. Osip'yan, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 661 (1989) [JETP Lett. **49**, 759 (1989)].  
 [6] C. L. Wang and G. Agnolet, Phys. Rev. Lett. **69**, 2102 (1992).  
 [7] E. Rolley, E. Chevalier, C. Guthmann, and S. Balibar, Phys. Rev. Lett. **72**, 872 (1994).  
 [8] R. M. Bowley and D. O. Edwards, J. Phys. (Paris) **44**, 723 (1983).  
 [9] P. Nozières and M. Uwaha, J. Phys. (Paris) **48**, 329 (1987).  
 [10] D. O. Edwards, S. Mukherjee, and M. S. Pettersson, Phys. Rev. Lett. **64**, 902 (1990).  
 [11] M. Faraday, Philos. Trans. R. Soc. London **121**, 319 (1831).  
 [12] See, e.g., J. Miles and D. Henderson, Annu. Rev. Fluid Mech. **22**, 143 (1990), for a recent review.  
 [13] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 851 (1993).  
 [14] A. B. Ezerskii, M. I. Rabinovich, V. P. Reutov, and I. M. Starobinets, Zh. Eksp. Teor. Fiz. **91**, 2070 (1986) [Sov. Phys. JETP **64**, 1228 (1986)].  
 [15] S. Douady and S. Fauve, Europhys. Lett. **6**, 221 (1988).  
 [16] S. Douady, S. Fauve, and O. Thual, Europhys. Lett. **10**, 309 (1989).  
 [17] S. Ciliberto, S. Douady, and S. Fauve, Europhys. Lett. **15**, 23 (1991).  
 [18] S. Fauve, K. Kumar, C. Laroche, D. Beysens, and Y. Garrabos, Phys. Rev. Lett. **68**, 3160 (1992).  
 [19] E. Bosch and W. van de Water, Phys. Rev. Lett. **70**, 3420 (1993).  
 [20] B. J. Gluckman, P. Marcq, J. Bridger, and J. P. Gollub, Phys. Rev. Lett. **71**, 2034 (1993).  
 [21] S. T. Milner, J. Fluid Mech. **225**, 81 (1991).  
 [22] S. Balibar *et al.* (unpublished).  
 [23] See, e.g., P. Nozières, in *Solids Far From Equilibrium*, edited by C. Godrèche (Cambridge University Press, Cambridge, 1991).  
 [24] L. D. Landau and E. M. Lifshitz, *Mechanics* (Pergamon, New York, 1978), Sec. 27.  
 [25] Actually, this is only true when the system is large enough that dissipation at the edges (where the surface can be pinned) may be neglected. See [13,21] for a more detailed discussion of damping mechanisms.  
 [26] This ceases to be true in the large damping regime; the effect of the anisotropy on the instability can then, however, be obtained only from a full numerical analysis of the Mathieu equation.  
 [27] This was suggested to us by G. Frossati.  
 [28] The electrically excited Faraday instability of a charged superfluid layer was observed by P. Leiderer, Physica (Amsterdam) **126B**, 92 (1984); ac magnetic fields can also be used in ferrofluids and mercury, see, e.g., V. G. Bashtovoi and R. E. Rosensweig, J. Magn. Magn. Mater. **122**, 234 (1993).  
 [29] Note, however, that manipulation of the shape of the crystal edge with the aid of electric fields may help clarify the effects of the boundary conditions on the formation of interface patterns — see [13] and references therein.