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Charge-Induced Instability of the ^{4}He Solid-Superfluid Interface

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The interface between crystalline and superfluid ^{4}He, when charged with negative ions, is deformed as a result of melting and crystallization processes. As long as the electric field normal to the interface is below a critical value, the deformation is stable and reversible. Above the critical field a charge-induced instability of the crystal is observed, which resembles the electrohydrodynamic instability of liquid surfaces.

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The boundary between crystalline and superfluid ^{4}He displays unique properties which have attracted much interest recently.[1-6] Among the unusual features of the quantum crystal surface is its similarity to the free surface of a liquid: Solid helium partially filling the sample cell forms a convex meniscus just like a liquid which does not wet the walls; this implies that certain portions of the solid-liquid boundary—depending on temperature and orientation—are atomically rough.[1,3,4] Furthermore, when the crystal surface is disturbed, the perturbation can propagate along the boundary as a result of rapid melting and crystallization processes, following a dispersion law much like capillary waves.[1,5]

In this Letter we report for the first time on the behavior of superfluid-solid ^{4}He interfaces charged with negative ions (i.e., electron bubbles). If this boundary were similar to a liquid surface the charges ought to lead to a loss of stability at high electric fields, analogous to the electrohydrodynamic instability as observed, e.g., on liquid ^{4}He and at the interface of phase-separated ^{3}He-^{4}He mixtures.[7-9] As our measurements show, such an instability—connected with the development of periodic interfacial deformations—does indeed occur, which adds to the intriguing character of this particular solid-liquid interface.

The experiments were performed in a cylindrical Plexiglas cell (inner dimensions: 20 mm high, 20 mm diam) with optical access both from the side and from the top. An electric field could be applied between the bottom and the top flanges, consisting of quartz plates with an evaporated gold mirror and a transparent conductive coating of In_2O_3, respectively. In order to determine the profile of the interface we have used the interference of light ($\lambda = 632.8$ nm) reflected from the two almost plane-parallel electrode plates,[8] a method which in principle is similar to the holographic technique of Avron *et al.*[3] Crystals were grown at constant temperature by slowly adding helium from a pressurized gas reservoir, until the solid filled the bottom of the sample cell up to a height of typically 4 mm. As in earlier experiments[3,5] crystals grown in this way showed no visible defects. The uncharged interface was completely flat, except for the rounding of the meniscus close to the walls of the cell. The temperature for all the measurements reported here was 1.35 K; the helium crystals therefore had hcp structure.

After a crystal was grown, the electrons for the charging of the interface were injected into the liquid by means of a field-emission tip, and drawn towards the solid phase by the applied electric field. To study the behavior of the electron bubbles at the liquid-solid boundary we started with a constant, relatively small amount of charge and observed the response of the interface as the field was varied. Figure 1 shows an example of an interferogram of such a charged boundary, together with the corresponding interfacial profile, for an applied voltage $U = 1400$ V and an estimated number of ions on the order of 10^6. Clearly a dimple has developed near the center of the cell, which is characterized by the following observations[10]: (i) For $U = \text{const}$ the dimple profile does not change even for times as long as 10^3 sec (provided U is below a critical value U_c). (ii) The profile is anisotropic, as a result of the anisotropy of the underlying crystal. (iii) When the applied field is varied, a new equilibrium shape of the interface develops with a time constant of about 20 sec. These deformations are reversible; they disappear completely when the electric field is switched off.

From the long-time stability at constant voltage we conclude that the interface presents a barrier for negative ions high enough to prevent the charges from penetrating from the liquid into the solid. This is supported by an estimate of the energy W_i of an ion which for the solid, because of its larger density, yields a value about 18 meV higher than for the coexisting liquid.[11] The relatively slow, nearly exponential change of the dimple depth when U is varied, on the other hand, is consistent with the observation of Keshishev, Parshin, and Babkin that melting-crystallization waves are strongly damped for $T \gtrsim 1$ K.[5]

In order to extend the theory of melting-crystallization waves[1] to charged interfaces we have included a term in the usual equation of motion which represents the pressure of the ions.[12] The resulting relation between the frequency ω and the wave vector q of these excitations is

$$\omega^2 = \frac{\rho_l}{\rho_s - \rho_l} g q + \frac{\rho_l}{(\rho_s - \rho_l)^2} \bar{\alpha} q^3 - \frac{i \omega q \rho_s \rho_l}{m_4 K (\rho_s - \rho_l)^2} - \frac{\rho_l}{4\pi (\rho_s - \rho_l)^2} q^2 (\epsilon_s E_s^2 + \epsilon_l E_l^2), \tag{1}$$

where we have assumed that the ions have a mobility large enough to warrant a constant electrostatic potential along the interface.[12] Here the subscripts s and l refer to the solid and the liquid, respectively; ρ denotes the density, g the acceleration due to gravity, ϵ the dielectric constant, m_4 the atomic mass of ^{4}He, and K the kinetic growth coefficient of the interface; $\bar{\alpha}$ is related to the interfacial tension α between liquid and solid He by $\bar{\alpha} = \alpha + \partial^2\alpha/\partial\varphi^2$ [φ is the angle between the normal of the displaced interface and the (vertical) z axis].[1]

An analysis of Eq. (1) shows that above a critical field solutions exist with a *negative* damping coefficient γ, i.e., these modes will grow instead of decaying in time. If we assume, for simplicity, that the interface is maximally charged ($E_1 = 0$), an instability is predicted to develop at a critical field $E_{s,c} = [64\pi^2 g \bar{\alpha}(\rho_s - \rho_l)/\epsilon_s^2]^{1/4}$ and a critical wave vector $q_c = [g(\rho_s - \rho_l)/\bar{\alpha}]^{1/2}$. The critical values, calculated with the material parameters for the hcp-superfluid interface at 1.35 K,[3] are $E_{s,c} = 1850$ V/cm and $q_c = 10.2$ cm^{-1}. When the field is raised above $E_{s,c}$ two features are expected: (i) Since $|\gamma|$ increases as well, the instability ought to develop more and more rapidly; (ii) the range of modes with negative damping (i.e.,

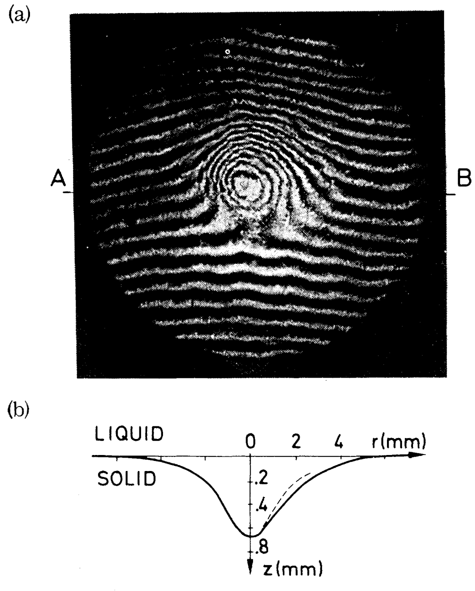

FIG. 1. (a) Interference pattern of an hcp-superfluid ^{4}He interface deformed by $\sim 10^6$ negative ions in an external field $E = 700$ V/cm, at a temperature $T = 1.35$ K. The parallel fringe pattern outside the center results from a slight inclination of the two interferometer plates. (The helium crystal in this region is flat, and hence the fringes are not distorted.) (b) Dimple profile determined from (a) along the direction A-B (full curve). A comparison with the dashed line, which is a mirror image of the left-hand profile, illustrates the asymmetry of the dimple.

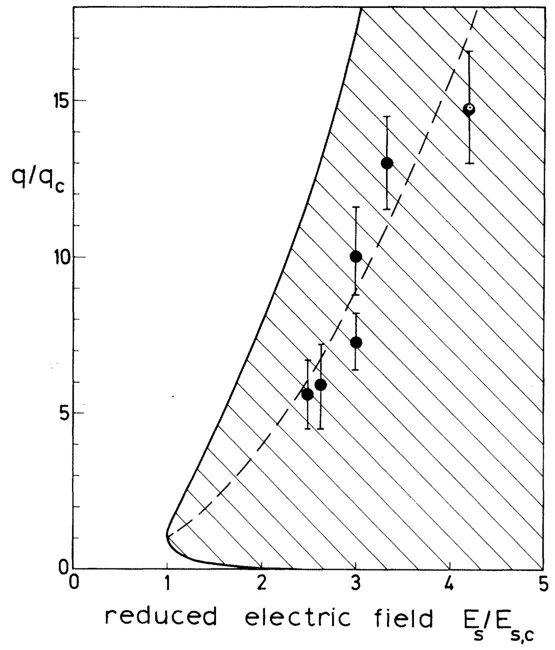

FIG. 2. Range of the charge-induced instability of the hcp-superfluid ^{4}He interface, indicated by the hatched area, as calculated from Eq. (1). The solid line is a plot of the reduced wave vectors q/q_c for which the damping coefficient vanishes; the dashed line indicates the unstable wave vectors with the largest gain. The experimental results, given by the circles, were obtained by determining the characteristic average wave vector q_m of unstable, corrugated structures like in Fig. 3.

gain) becomes larger, and hence the deformation profile of the interface will contain a wider and wider spectrum of Fourier components. This is illustrated in Fig. 2, where the range of unstable wave vectors is plotted versus the reduced electric field $E_s/E_{s,c}$.

An experimental example for an hcp-superfluid interface at supercritical conditions is shown in Fig. 3. A voltage of 1800 V was applied between the electrodes 1 sec before the picture was taken. Since the ion source was switched on continuously, the interface, because of the large mobility of the ions in superfluid ^{4}He,[13] was charged close to saturation. For a crystal height of 4 mm this implies an ion density $n = 2.7 \times 10^9$ cm^{-2} and a corresponding reduced electric field $E_s/E_{s,c} = 2.4$. As the picture demonstrates, corrugations appear consisting of a large number of small dimples with a tendency toward parallel alignment (although the influence of the cylindrical geometry is still visible). The dimples increase in depth, until finally, after a few seconds, charged liquid bubbles split off from the dimple tips and move through the solid at a velocity of 1 mm/sec. Related phenomena at the gas-liquid interface of ^{4}He have been described by Volodin, Khaikin, and Edel'man.[14]

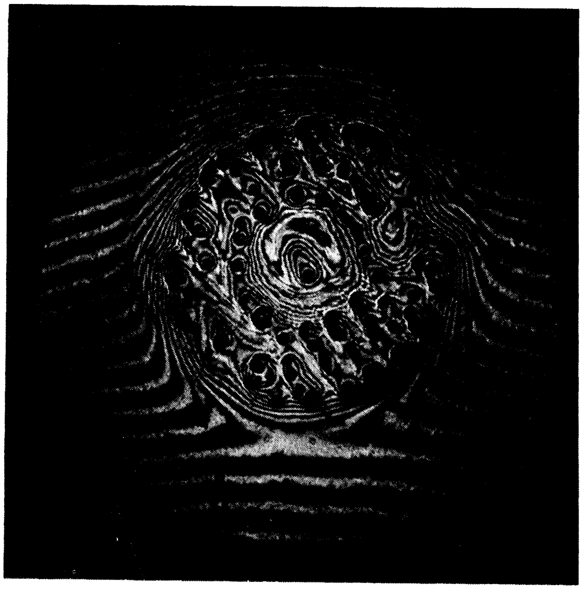

FIG. 3. Interference pattern of an hcp-superfluid ^{4}He interface undergoing a charge-induced instability, about 1 sec after a (supercritical) voltage U = 1800 V was applied between the two electrodes. An additional holding field resulting from negative charges on the cylindrical cell walls confines the ions on the crystal surface in a circular central patch about 1 cm in diameter.

The higher the applied voltage the faster this instability is found to develop (in ~ 50 sec at 1550 V, ~ 1 sec at 3000 V); simultaneously the wave vector q_m characterizing the spacing of the corrugations becomes larger and less well defined. Results for several electric fields are plotted as circles in Fig. 2. All these observations are in close agreement with the predictions derived from Eq. (1).

The direction of the stripes in Fig. 3 apparently depends on the anisotropy of the helium crystal. Although the crystal orientation has not been determined here, we have recorded the facets which appear during rapid crystal growth even above the supposed roughening transition near 1 K.[3-5] The corrugations resulting from the instability preferentially develop parallel to the plane of the largest facet (even though in these experiments the interface was only charged in thermodynamic equilibrium, long after all the visible facets had disappeared).

In summary, we have observed a charge-induced instability of the ^{4}He solid-superfluid interface analogous to the electrohydrodynamic instability of liquid surfaces. In contrast to the previously investigated liquids, however, here the excitations of the interface—melting-crystallization waves—are strongly damped. Furthermore, the present measurements extend the instability studies, which so far had concentrated on the regime $E/E_c \sim 1$ and $q/q_c \sim 1$, deep into the unstable region.

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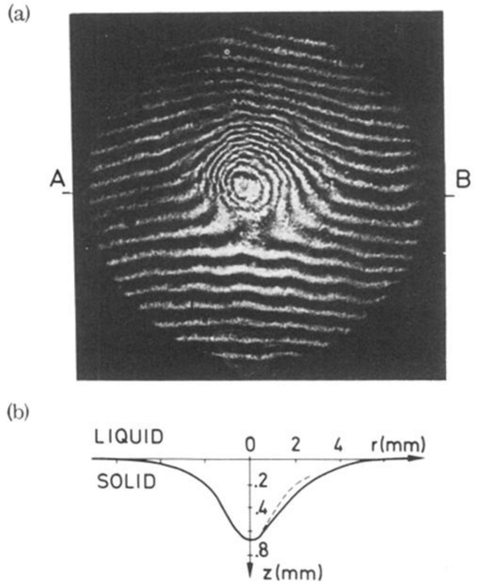

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