Stress-Induced Melting and Surface Instability of ^4He Crystals.

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Abstract. – The response of the interface between liquid and solid ^4He is investigated when the helium crystal is exposed to uniaxial stress. It is observed that part of the crystal melts, the change in height of the interface being roughly proportional to the square of the stress. Beyond a critical value the interface becomes unstable and develops corrugations with a wave vector of the order of the inverse capillary length, which is associated with the so-called Grinfeld instability. These results confirm recent predictions by Balibar, Edwards and Saam.

When a uniaxial stress is applied to a crystal with a constant mass, its cross-section in the direction perpendicular to the applied force usually grows, the magnitude of this effect being given by the Poisson number. However, if the crystal is in thermodynamic equilibrium with its melt, the opposite should happen, as pointed out by Balibar, Edwards and Saam [1]. On uniaxial compression part of the crystal ought to melt, and for sufficiently large strain the crystal surface should even display an instability, showing up as a spontaneous deformation, the Grinfeld instability [2].

In this paper we report first experiments on the observation of these effects. The system investigated here was ^4He around 1 K, which for several reasons has already proved very suitable for studying phenomena at the crystal-melt interface [3-6]. The measurements reveal the partial melting of a helium crystal caused by external stress generated by a piezoelectric transducer, an effect which occurs both upon uniaxial compression and expansion. We also demonstrate the appearance of corrugations at the liquid-solid interface for large stress, with a characteristic wave vector on the order of the inverse capillary length

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Fig. 1. – Sketch of the sample cell: a) side view; b) bottom view; m, mirror, w, window; p, piezoelectric bimorph; f, metallic frame.

of the interface. Furthermore it is shown that corrugations of the liquid-solid ^4He interface reported in an earlier paper [6] for a crystal during cooling—then tentatively ascribed to an instability caused by heat flow—are actually also due to crystal strain (resulting from the pressure-dependent molar volume of He crystals at the coexistence curve). A preliminary report of these results has already been given earlier [7].

The experimental cell (see fig. 1) was similar to the one used earlier, built as an interferometer with optical access from the bottom of the cryostat. Local changes of the crystal height could thus be detected via a shift of interference fringes with a vertical resolution of about one μm . In addition, a piezoelectric bimorph transducer mounted in a rectangular frame allowed us to stress the helium crystals in the two sections (A and B) of the frame in a horizontal direction. The parts of the crystal outside the frame (part C) were not directly affected by the piezo and served as a reference. In a typical run, the crystal was grown to a height of about 4 mm, covering the whole bottom plate of the cell with a diameter of 30 mm. Little holes at the bottom of the frame between the various sections provided a coupling during crystal growth [8], so that the crystal orientation was the same in all parts of the cell. The orientations investigated here were random (the basal plane of the He crystal was in no case horizontal), which implies that the crystal surface was atomically rough in our temperature range [3].

An important quantity for this experiment is the absolute value of the stress exerted on the helium crystal by the piezoelectric transducer plate. Since it is known that the elongation of piezoelectric ceramics is strongly reduced at low temperature, first the bending motion of the transducer plate was measured in liquid helium as a function of the applied voltage by means of a microscope with large working distance. A typical response of the free transducer end was $dx/dV = 80 \mu\text{m}/100 \text{ V}$. With the He crystal being grown to the characteristic height, the transducer end was essentially clamped by the solid, so that its motion was below the optical resolution of $3 \mu\text{m}$. The force exerted by the transducer upon applying a voltage V can then be calculated from

$$F = \frac{dF}{dx} \frac{dx}{dV} V. \quad (1)$$

The «spring constant» dF/dx of the transducer plate was measured at room temperature and extrapolated to low T using the known temperature dependence of the elastic constants of the plate materials.

It was shown previously that one can partially melt a He crystal by applying a vertical stress on the solid, but not on the coexisting liquid phase [6, 9]. In these earlier measurements the pressure was directed perpendicular to the liquid-solid interface, generated by electron bubbles which were pulled against the interface under the influence of

Fig. 2. - Height change h of a ^4He crystal under uniaxial stress. In a), b) positive stress is applied at time $t = 0$ and switched off at $t = 21$ s. The two curves are for transducer voltages of 15 (a)) and 20 V (b)). In c), a voltage of -20 V is applied to the transducer at $t = 0$, resulting in negative strain.

an applied electric field. As a result the crystal melted by an amount proportional to the applied electrostatic pressure.

In contrast to this linear effect a quadratic response is expected if the crystal is subjected to a nonhydrostatic strain, as in the present experiment. According to Balibar, Edwards and Saam [1] one expects a depression

$$h = - \frac{(1 - \sigma_p)}{\Delta \rho g} \frac{1}{E} (\delta \sigma)^2. \quad (2)$$

Here h is the height change of the crystal, $\sigma_p = 0.33$ is Poissons's ratio, $E = 3.05 \cdot 10^8 \text{ erg/cm}^3$ is Young's modulus, $\delta \sigma$ is the stress, $\Delta \rho$ is the density difference between liquid and solid and g is the acceleration due to gravity.

In our measurements we have tried to check this prediction experimentally. Results for the height change of a ^4He crystal under stress are presented in fig. 2 for a temperature of 1.0 K. The time to reach the new equilibrium after changing the applied stress was about 3 seconds, in agreement with the known relaxation time of the solid-superfluid ^4He interface at this temperature [6]. In fig. 2a) and b), which show experimental traces for the crystal height near the centre of section A, the voltage was applied to the transducer at time $t = 0$, leading to a compression of the He crystal. At $t = 21$ s the voltage was switched off, and the crystal grew back to its original height. Figure 2c) shows the response of the same part of the

Fig. 3. – Height change h for two different ^4He crystals vs. the voltage applied to the piezoelectric transducer (□ crystal 1, ◇ crystal 2). A voltage of 1 V corresponds to a stress of $1.54 \cdot 10^{-2}$ bar. The solid line represents a quadratic dependence of h on the applied stress, the absolute value of h in this case being about a factor of 3 larger than that given by eq. (2).

crystal, but now with the sign of the transducer voltage being reversed, which then gives rise to a *dilatation* of the crystal at $t = 0$. Also in this case the crystal height was reduced, pointing towards the expected quadratic effect. (The absolute value of h was smaller for the expansion than for the compression under otherwise similar conditions, possibly due to details of the coupling between the transducer surface and the He crystal). In fig. 3 we have plotted data for h at several values of the transducer voltage (compression in this case), which show a nearly quadratic dependence of h on the crystal stress indeed. Also the absolute value of the effect is of the order of magnitude predicted by eq. (2). It has to be mentioned that the measurements often were hampered by various problems, like apparent inhomogeneities in the stress at different parts of the crystal, which led to a position-dependent response, large fluctuations in the crystal height and a gradual reduction of the response occurring when a large stress was repeatedly applied to a crystal. More detailed experiments are therefore required. Nevertheless our results clearly demonstrate the existence of the predicted melting of strained crystals at least on a qualitative level.

A related, yet even more striking phenomenon is the instability of the crystal surface when the stress is increased beyond a critical value. This has been discussed by Grinfeld [2] and later by Nozières [10] and Balibar, Edwards and Saam [1], who calculated the critical stress $\delta\sigma_c$ to be given by

$$\delta\sigma_c^2 = \frac{(\gamma \Delta\rho g)^{1/2} E}{(1 - \sigma_p^2)}. \quad (3)$$

The instability, being opposed by an increase in the gravitational energy and surface energy at small and large wave vectors, respectively, should appear as a corrugation of the interface with a spacing equal to $2\pi a$, where $a = (\gamma/\Delta\delta g)^{1/2}$ is the capillary length. Since the instability threshold is lowest in the direction with the smallest surface stiffness γ , the wave vector of the spontaneous corrugations should point in this direction. For ^4He around 1 K

under consideration here the calculated critical values are [1]

$$q_c = \frac{1}{a} = 9.4 \text{ cm}^{-1} \quad \text{and} \quad \delta\sigma_c = 2.5 \cdot 10^{-2} \text{ bar}. \quad (4)$$

In our experiment also this predicted instability could be observed, an example being shown in fig. 4. The voltage applied to the transducer was in this case 200 V which generates a stress of $3.1 \cdot 10^{-2}$ bar, well above the instability threshold. The dark stripes, marked by the arrows, indicate grooves in the crystal with a depth of about one mm, developing spontaneously on a time scale of seconds. The average spacing was 4 mm, somewhat below the value of 6.7 mm expected according to eq. (4). These corrugations were not permanent, but gradually disappeared within a few minutes (even when the transducer voltage was kept constant), probably due to a reduction of the crystal strain by the motion of defects and/or vacancies. The fact that the wave vector of the corrugation is larger than the theoretical value at the instability point may arise from «quenching» the system far into the unstable regime, where the wave vectors of the modes with the fastest growth, which dominate the instability, are expected to increase (similar to the electrohydrodynamic instability of the charged solid-superfluid interface of ^4He [9]).

Finally, we would like to discuss the observation of Bodensohn *et al.* [6] that cooling a ^4He crystal at a temperature of about 1 K also led to an instability, whose appearance was quite similar to the one described above. It has been pointed out by Balibar *et al.* [1] that this instability might also have been caused by stress, because for $T \geq 1$ K a variation in temperature is accompanied by a considerable change in molar volume mainly as a consequence of the change in pressure along the melting curve [11]. Originally Bodensohn *et al.* had suggested that heat flow across the crystal-melt interface could also give rise to an instability. It was shown by Bowley and Nozières [12], however, that although this heat flow mechanism should in principle exist, it would be far too small to account for the observed effect in the case of the solid-superfluid interface of ^4He because of the extremely effective thermal transport in the superfluid phase. In order to clarify whether also the thermally induced instability is basically of the Grinfeld type we have investigated the conditions under

Fig. 4.

Fig. 4. – Instability pattern of the solid-liquid ^4He interface. The indentations, which appear as dark stripes in sections A and B, are additionally marked by arrows.

Fig. 5.

Fig. 5. – Instability pattern of the solid-liquid ^4He interface, appearing at a temperature of 1.18 K on a crystal which was grown at 1.09 K. Indentations in this case are white. The scale bar marks a length $2 \text{ pa} = 6.7 \text{ mm}$.

TABLE I. - Dependence on heating rate. t : time for increasing the temperature from T_0 to T_{inst} .

T_0	T_{inst}	t [s]
1.09	1.18	40
1.09	1.18	91
1.09	1.19	175
1.09	1.19	308
1.09	1.18	600
1.09	1.18	1212

which a change in temperature, rather than a change in piezo voltage, leads to interfacial corrugations. An example of such a pattern appearing when a ^4He crystal is cooled is shown in fig. 5. As before, the spacing of the corrugations is close to $2\pi a$. It was found that starting with a crystal grown at a temperature T_0 the instability developed either when the temperature was decreased or raised. For the latter case we have listed in table I the temperature T_{inst} where the corrugations first appeared. The growth temperature was 1.09 K, and the heating rate was varied between 0.074 and 2.25 mK/s. Within the accuracy of the determination of T_{inst} , 10 mK, the temperature difference $\Delta T = T_{\text{inst}} - T_0$ was found to be constant, irrespective of the heating rate and hence of the heat flow across the interface. Consequently the instability cannot be caused by thermal transport, in agreement with the result of Bowley and Nozières.

Strong support to the suggestion by Balibar *et al.* that the instability induced by a temperature change is in fact due to strain in the crystal is lent by the results listed in table II. The temperature difference between the instability onset T_{inst} and the starting temperature T_0 decreases strongly as T_0 is raised. This is exactly what is expected on the basis of the rapidly increasing slope of the melting curve above 1 K and the corresponding change in the molar volume of the solid [11]. As the last column in table II shows, the instability always starts to develop when a certain value of the change in molar volume, $\Delta V \approx 5 \cdot 10^{-3} \text{ cm}^3/\text{mole}$, and hence a certain (in this case negative) critical strain is reached. The experimentally determined values of ΔT given in table II agree very well with the estimates quoted in the work of Balibar *et al.*

As observed already by Bodensohn *et al.* the direction of the wave vector of the ΔT -induced instability pattern was parallel to the projection of the c -axis of the helium crystal along the liquid-solid interface (which can be identified during the crystal growth, because it is the direction with the smallest growth coefficient [6]), indicating that this is the «easy axis» for the formation of the instability.

We should add that for very small heating rates one should eventually expect larger temperature differences $T_{\text{inst}} - T_0$, because part of the strain developing due to the change in molar volume will relax by the diffusion of vacancies.

In summary, our results verify the main predictions for the response of a solid-melt

TABLE II. - Appearance of the instability.

T_0	T_{inst}	ΔT [K]	$\Delta V [10^{-3} \text{ cm}^3/\text{mole}]$
1.00	1.12	0.12	5.4
1.09	1.18	0.09	6.0
1.18	1.23	0.05	5.3
1.24	1.27	0.03	3.9

interface when the solid is under nonhydrostatic external stress. Both partial melting of the crystal and the Grinfeld instability of the interface have been observed. These findings, reported here for the quantum system ^4He , should occur for classical crystals as well, and therefore are of general relevance.

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