

Measurements on the Surface Tension of ^3He Crystals near 100 mK

Igor A. Todoshchenko, Harry Alles, Heikki J. Junes,
Alexander Ya. Parshin*, and Viktor Tsepelin†

*Low Temperature Laboratory, Helsinki University of Technology
P. O. Box 2200, FIN-02015 HUT, Finland*

**P. L. Kapitza Institute, Kosygina 2, Moscow 119334, Russia*

†Department of Physics, Stanford University, Stanford, CA 94305-4060, USA

^3He crystals start to show facets on their surface only at about 100 mK, well below the roughening transition temperature, and the reason for this change of the surface state is not clear yet. However, the most important characteristic of the crystal surface, the surface tension, was not measured in this temperature range before. We report our observations on the equilibrium shape of the ^3He crystals in the temperature range of 77...110 mK. The surface tension was found to be isotropic and temperature-independent, and the corresponding value of the capillary length, $\lambda = 0.93 \pm 0.10$ mm, is in a good agreement with the value measured at higher temperatures by Rolley et al.²

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1. INTRODUCTION

The solid-liquid interface of helium is a very interesting physical object. It exists down to very low temperatures where the two bulk phases provide effective transport of mass and heat, resulting in high mobility of the crystal surface. This gives a unique opportunity to investigate the intrinsic properties of the interface which can thus serve as a good model system for surface physics. In addition, due to the large amplitude of zero-point motion of atoms, the surface of helium crystals is essentially a quantum system.

One of the most exciting phenomena on the crystal surface is faceting. Facets are smooth planes in the high-symmetry directions which appear due to the pinning of the interface to the underlying lattice. However, thermal fluctuations of the interface suppress the pinning when temperature increases

and, as a result, facets disappear at a certain temperature. The amplitude of thermal fluctuations is limited by the surface energy, and consequently the roughening temperature is directly related to the surface tension α : $T_R = (2/\pi)\alpha d^2$, where d is the height of an elementary step on a facet.¹

Rolley *et al.*² have measured the surface tension of ^3He crystals at 140... 330 mK and found it to be isotropic and temperature-independent. The obtained value of the surface tension, $\alpha = 0.060 \pm 0.011$ erg/cm, yields $T_R = 260 \pm 50$ mK, or more than twice higher than the temperature at which facets have been observed in the experiment, $T_{obs} \approx 100$ mK. Iordanskii and Korshunov³ have shown that quantum fluctuations of the interface cannot reduce the transition temperature, but provide the exponential decrease of the free energy of an elementary step, and facets may be simply too small to be observed. Thus, the fact that facets appear at 100 mK is not related to the roughening transition, but is a manifestation of an increase of the step energy in this temperature range. The value of the step energy β is set by the surface tension and the energy of the pinning V : $\beta = 4d/\pi \cdot \sqrt{\alpha V}$.⁴ In this work we present the first measurements on the surface tension of the bcc ^3He crystals near 100 mK.

2. EXPERIMENTAL METHODS

The most natural way to obtain the surface tension is to measure the equilibrium shape of a free crystal surface. For measurements on the surface tension, the crystal size should be larger than the supposed capillary length $\lambda \sim 1$ mm, otherwise the equilibrium shape will be nearly spherical as in the absence of gravity, and will not provide any means to obtain the surface tension. In the case of ^3He at temperatures near 100 mK, however, it is extremely difficult to grow a large single crystal because of the large latent heat and poor thermal conductivity of the liquid. During growth the crystal becomes colder than the liquid far away from the crystal, and the equilibrium melting pressure $p_m(T)$ on the interface becomes higher than near the cell walls (the slope of the melting curve of ^3He is negative below 320 mK). If this difference in the melting pressures exceeds the threshold for the crystal nucleation, $\delta p \sim 3$ mbar, a new crystal appears and grows instead of the existing one. The critical growth rate for such a process is $v_{nucl} \sim 0.1 \mu\text{m/s}$.⁵ A comparable restriction is imposed by the Mullins–Sekerka instability of the solidification front, which results in appearance of "fingers" on the moving surface.⁶ These restrictions imply that growing of a high quality crystal of the size of a few millimeters takes at least 10 hours.

We have carried out our experiment in the compressional cell, pressur-

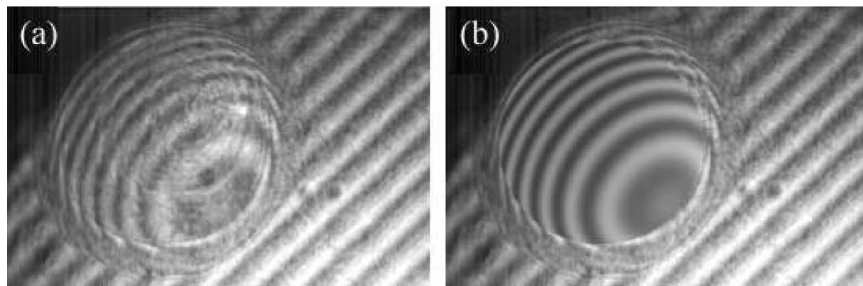


Fig. 1. (a) Interference pattern of a relaxed ^3He crystal at 102 mK. The size of the imaged area is $6.2 \times 4.8 \text{ mm}^2$. (b) Generated pattern of the corresponding equilibrium crystal shape [solution of Eq. (1), see text] inserted to it.

ized by ^4He from room temperature. In order to have very small controllable compression rates and to avoid fluctuations of the pressure in the ^4He filling line, we have mounted a cryogenic valve on the mixing chamber plate. Crystals were nucleated by pressurizing liquid ^3He above the equilibrium melting pressure. In order to create the crystal seed in the field of view, we applied a high voltage to a sharp tip placed near the boundary of the illuminated area. The crystal were imaged with a low temperature Fabry-Pérot interferometer.⁷ The interference pattern [see Fig. 1(a)] was projected to the CCD sensor working at 65 K. The sensor has 383×575 pixels, each corresponding to an area of $15 \times 15 \mu\text{m}^2$.

After the crystal has been carefully grown to the required size, it has to relax in order to obtain the equilibrium shape. There are two relaxation times, one is to reach the thermal equilibrium in the experimental cell after growing, τ_{th} , and another one is to reach the phase equilibrium on the crystal surface, τ_{ph} , both relaxation times being rather long. The time constant of thermal relaxation is determined by the heat conductivity and the heat capacity of liquid ^3He , $\tau_{th} \sim CD^2/\kappa \sim 1$ hour for the cell of the size $D \sim 1$ cm. In practice this means that even if one grows the crystal very carefully, so that the variation of the equilibrium melting pressure $p_m(T)$ inside the cell does not exceed 1 mbar, one should still wait about 10 hours after growing until this variation is much smaller than the characteristic hydrostatic pressure of $\sim 10 \mu\text{bar}$ which drives the relaxation of the surface.

The time τ_{ph} can be evaluated as follows: the hydrostatic pressure $\delta p_h \sim \rho_l gh$ drives the motion of the interface with the velocity $v \sim h/\tau_{ph} \sim k\delta p_h \cdot \Delta\rho_{sl}/(\rho_s\rho_l)$, where $k(100 \text{ mK}) = 2.5 \cdot 10^{-7} \text{ s/cm}$ is the effective growth coefficient of the interface,⁸ and one finds $\tau_{ph} \sim \rho_s/(\Delta\rho_{sl}gk) \sim 10^5 \text{ s}$. During the phase relaxation it is important to keep the temperature in the experimental cell constant with high accuracy since the change in temperature

results in total growth or melting of the crystal. To stabilize the temperature, we have used a feedback program which operates on the heater attached to the mixing chamber and uses integrated signal from the mechanical pressure gauge to control the melting pressure $p_m(T)$. We were able to keep the value of the melting pressure with accuracy better than $1 \mu\text{bar}$.

Another experimental difficulty arose from the fact that we were not able to close the ^4He filling line completely, thus the total amount of solid ^3He inside the cell slightly varied with time. However, in order to reach the equilibrium with needed accuracy, the growth rate of a crystal surface due to compression (decompression) of the cell, v_c , should be much smaller than the growth rate due to phase relaxation process, v_{rel} . Our cryogenic valve allows to apply the compression (decompression) rates \dot{V} as small as $10^{-10} \text{ cm}^3/\text{s}$. This corresponds to the average growth (melting) rate of a crystal $v_c = \dot{V}(\rho_s/\Delta\rho_{sl})/S \approx 2 \cdot 10^{-8} \text{ cm/s}$ for a typical surface area $S \approx 20 \text{ mm}^2$. We can follow the relaxation process until the crystal surface becomes as close to the equilibrium shape as our vertical resolution $\Delta h \approx 20 \mu\text{m}$. Even at this stage the velocity of the relaxing surface, $v_{rel} \sim k\Delta hg\Delta\rho_{sl}/\rho_s \sim 3 \cdot 10^{-8} \text{ cm/s}$, is still higher than the velocity due to the slow compression (decompression) of the cell. Thus, the effect of slow compression of the cell is negligible. In fact, we have done our experiments mostly on slowly melting crystals in order to avoid appearance of facets below 100 mK.

With all precautions enumerated above we were able to grow a single ^3He crystal with a size of a few millimeters and to follow the relaxation of its surface to the equilibrium shape at several temperatures in the range of 77 ... 110 mK. At all temperatures, the observed crystal shapes were perfectly axisymmetrical. We conclude that, within the accuracy of our measurements, α remains isotropic down to 77 mK.

3. DATA ANALYSIS AND RESULTS

If the surface tension is isotropic, the equilibrium crystal shape (ECS) in the gravitational field obeys the Laplace equation $g\Delta\rho_{sl}z(x, y) + \alpha K(x, y) = \text{const}$, where $K(x, y)$ is the local curvature of the surface. For the crystal lying on the horizontal plane, ECS is cylindrically symmetric, and the Laplace equation reduces to the second-order ordinary differential equation

$$\frac{r''_{zz}}{(1 + (r'_z)^2)^{(3/2)}} - \frac{1}{r(1 + (r'_z)^2)^{(1/2)}} + \frac{1}{\lambda^2}(z - z_0) = 0, \quad (1)$$

where r is the radius of the crystal at height z , $\lambda = \sqrt{\alpha/(g\Delta\rho_{sl})}$ is the capillary length, and z_0 is the constant which determines the volume of

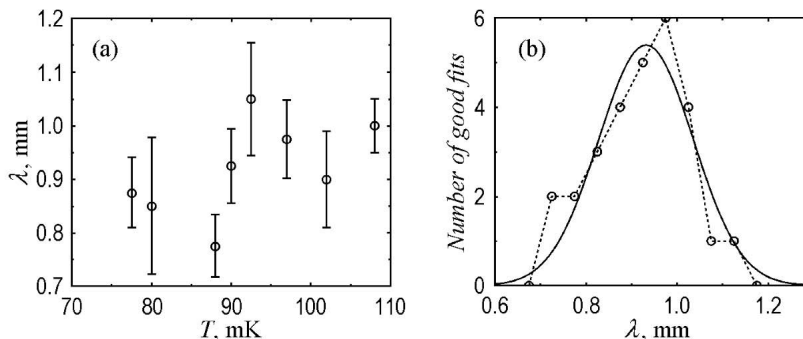


Fig. 2. (a) Measured values of the capillary length. (b) Histogram of good fits (see text).

the crystal. We have set the boundary conditions at the vertical edge of the crystal, whose height was chosen as a reference: $r(0) = r_{max}$, $r'_z(0) = 0$. The solution of Eq. (1) can be found numerically, λ and z_0 being free parameters. For any given λ and crystal diameter $2r_{max}$, there exists only one value of z_0 , which ensures that the solution satisfies another boundary condition at the top of the crystal, $z'_r(r = 0) = 0$. Thus, with a given crystal diameter, we have only one fitting parameter, the capillary length λ , to describe the crystal shape observed in the experiment.

In fact, below the roughening transition temperature the surface tension is not isotropic, but has cusps in the directions perpendicular to the facets, and these cusps are responsible for the appearance of facets on ECS. However, near 100 mK the equilibrium size of a facet on ^3He crystals is very small. Landau⁹ has shown that the length of a facet L is proportional to the step energy β : $L \approx \beta R / (\alpha d)$, where R is the crystal radius. Rolley *et al.*² have estimated the step energy at 100 mK to be smaller than 10^{-11} erg/cm, which gives $L < R/200 \sim 10 \mu\text{m}$, or smaller than the size of the area imaged by a single pixel of our CCD sensor. Indeed, after long relaxation our crystal had a nice symmetric shape [see Fig. 1(a)], from which we concluded that (with the accuracy we have) the relaxation is completed and the surface tension is isotropic.

The data was treated as follows. At first stage, we measured the diameter of the crystal, $2r_{max}$, and found the axis of symmetry by marking the points on the crystal edge and fitting them with a circle. At the second stage, we fixed some trial value of the capillary length λ and found the value of the parameter $z_0 = z_0(r_{max}, \lambda)$ which provided that the solution of Eq. (1) satisfied the boundary condition at $r = 0$. Then we generated the interference pattern corresponding to the obtained profile [Fig. 1(b)] and compared it with the original intensity distribution [Fig. 1(a)]. Repeating this proce-

dure from the second stage with different λ , the range of the capillary length values which yielded good fits was obtained.

The results are shown in Fig. 2(a). Despite the scatter of the data, we can affirm that there is no remarkable variation in the value of the capillary length with temperature. Moreover, within the experimental error, our results coincide with the value 1.03 ± 0.09 mm obtained at temperatures above 140 mK by Rolley *et al.*² To find the best value of the capillary length and to estimate the error bar, we have plotted all values which yielded good fits at different temperatures as a histogram and fitted it with Gaussian distribution [see Fig. 2(b)]. In this way we have found that $\lambda = 0.93 \pm 0.10$ mm, which corresponds to the value of the surface tension $\alpha = 0.052 \pm 0.011$ erg/cm².

4. CONCLUSIONS

We have performed the first measurements on the surface tension of the bcc ³He crystals in the temperature range of 77...110 mK. With our experimental accuracy we did not observe any increase of the surface tension below 100 mK, where facets start to appear on the crystal surface. We can thus conclude that faceting of ³He crystals does not relate to the change in the value of the surface tension but rather to the enhancement of the pinning of the solid-liquid interface to the crystal lattice.

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