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Author(s): Babkin, A. V. & Alles, H. & Hakonen, Pertti J. & Parshin, A. Ya. & Ruutu, J. P. & Saramäki, J. P.

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## Observation of a New Surface State on $^4\text{He}$ Crystal Interfaces

A. V. Babkin, H. Alles, P. J. Hakonen, A. Ya. Parshin,\* J. P. Ruutu, and J. P. Saramäki

*Low Temperature Laboratory, Helsinki University of Technology, 02150 Espoo, Finland*

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The equilibrium shape of hcp  $^4\text{He}$  crystals has been studied at temperatures  $0.05 \leq T \leq 0.7$  K by means of a high-precision optical interferometer. We find that the profile of the interfacial boundary, close to an almost horizontal  $\mathbf{c}$ -facet, has a well-defined slope discontinuity separating two angular regions with different behavior of the surface stiffness  $\tilde{\alpha}$ . For surfaces tilted by an angle  $\phi \leq 100$   $\mu\text{rad}$  with respect to the  $\mathbf{c}$ -facet, we obtain  $\tilde{\alpha} = \kappa/\phi$ , contrary to the linear angular dependence predicted by current theories. Our results on four large crystals yield  $\kappa = (11 \pm 3) \times 10^{-4} T$   $\text{erg}/\text{cm}^2 \text{K}$ .

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Owing to the discrete nature of a lattice, the microscopic structure of a crystalline interface is influenced by its crystallographic orientation, creating a variety of possible surface states, which is reflected in the details of the equilibrium crystal shape (ECS). For example, the ECS may contain planar facets joined by curved interfacial regions, indicating a coexistence of two surface phases: atomically smooth and atomically rough. The transition region between a smooth facet and an adjacent curved part of a crystal boundary, also known as a *vicinal* interface, is an intermediate case between atomically smooth and rough phases. A vicinal surface, inclined by a small angle  $\phi \ll 1$  with respect to a high symmetry face, can be considered as a set of atomically smooth terraces separated by elementary steps. The thermodynamics of these steps determines the surface energy  $\alpha(\phi)$  and its signatures in the ECS. During the last decade, different theoretical models of the vicinal state have been considered [1]. Owing to very rapid relaxation at the liquid/solid interface [2], the most detailed experimental studies so far have been performed on  $^4\text{He}$  crystals [3,4].

Strong support of terraced vicinal planes [5] has been obtained in the most recent experiments by Rolley *et al.* [6]. This description of the interface leads to detailed predictions of the ECS close to a facet: the profile should show a power-law ( $x^{3/2}$ ) dependence. We have made high-resolution interferometric investigations of the crystal profile at small interfacial angles  $\phi \leq 10$  mrad with respect to the (0001) basal plane ( $\mathbf{c}$ -facet). In this Letter we present experimental data which show that power-law behavior, observed at  $\phi \geq \phi_{c2} \approx 2$  mrad, changes discontinuously into exponential relaxation at  $\phi \leq \phi_{c1} \approx 0.2$  mrad. This discontinuous change in the orientation of the interface is a clear indication of a first-order phase transition on the surface. The novel surface state at  $\phi \leq \phi_{c1}$  is characterized by a strong,  $1/\phi$ -dependent surface stiffness, which is difficult to explain within the framework of current theoretical models.

The experimental apparatus is a modification of an accurate optical interferometer, used in our previous observations on the free surface of superfluid  $^3\text{He}$  [7,8].

In our experiments we used regular commercial  $^4\text{He}$  gas with  $\leq 100$  ppb of  $^3\text{He}$  impurities. The  $^4\text{He}$  crystals were grown inside a carefully polished conical copper cell, illustrated schematically in Fig. 1. The body of the cell was thermally linked to the mixing chamber of a small dilution refrigerator, capable of cooling samples to  $\approx 40$  mK. In equilibrium, a crystal fills the entire conical section of the vessel: the lateral size of the interfacial region for all our samples was 35–40 mm in diameter. The  $\mathbf{c}$ -facet was aligned almost parallel to the upper part of the fused silica optical wedge ( $\Delta\phi \leq 10$  mrad), by the procedure described in Ref. [9]. The crystals were grown from 1-mm-diameter seeds at  $T = 50$  mK using an interfacial growth rate of  $\leq 0.2$   $\mu\text{m}/\text{s}$ . When the desired crystal size was reached, the filling capillary was closed with a low temperature valve.

The central part of the crystal is illuminated from above with a collimated 5-mm-diameter He-Ne laser beam. Light reflected from the interface and from the reference flat form an interferometric image of the crystal boundary onto a cooled charge coupled device (CCD) sensor. Both the bellows-operated focusing system and the CCD imager are located inside the 4 K vacuum can, close to the experimental sample. Compared with our

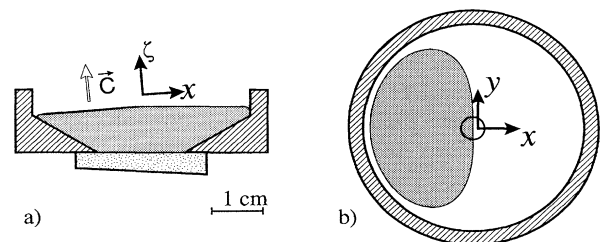


FIG. 1. Schematic drawing of the experimental cell. (a) Under equilibrium the crystal (dark shading) rests on the optical reference flat (light shading), which also serves as the bottom window of the experimental cell. The  $\mathbf{c}$ -facet of the sample is inclined by a small angle  $\tau = 3\text{--}15$  mrad with respect to the gravitational horizon. (b) Top view of the experimental container. The small circle in the center outlines the region of observation. The shaded area corresponds to the facet.

previous imaging system [7], the sensitivity to light has been significantly improved by employing an astronomy-grade slow-scan CCD camera as in Ref. [10]. The careful design of our interferometer, together with advanced fringe analysis methods, provide a vertical resolution of 5 nm. Details of the apparatus will be published elsewhere [11].

The lateral size of a facet is strongly influenced by its orientation with respect to the gravitational horizon: the size is at maximum for an exactly horizontal facet, whereas it progressively shrinks with increasing inclination. Therefore, the relative position of the facet edge within the field of observation [Fig. 1(b)] could be adjusted simply by tilting the entire cryostat slightly ( $\leq 20$  mrad). Since the crystal diameter is much larger than the capillary length of solid helium ( $a \approx 1$  mm), the curvature of an interface along the facet border, located in the center of the cell, is exponentially small.

Figure 2 displays the crystal profile in the edge region for one of our samples at 0.51 K. It is clearly seen in the magnified view [Fig. 2(b)] that the crystal shape actually contains a slope discontinuity, from  $\phi_{c1} = 430$   $\mu$ rad to  $\phi_{c2} = 2.3$  mrad, which separates two regions characterized by distinctly different behavior. At  $\phi \leq \phi_{c1}$ , we find  $\zeta(x) \propto \exp(x/x_0)$ , where  $\zeta(x) \leq 0$  describes the profile in a coordinate frame corresponding to the c-facet plane [see Figs. 1(a) and 2] and  $x_0$  is a fitting parameter. At  $\phi \geq \phi_{c2}$ , the profile is of the form  $x^\theta$  where  $\theta = 1.3-1.8$ ; the large span of  $\theta$  is caused by the uncertainty of the actual starting point for the power law.

In our one-dimensional case, the equation describing the ECS under gravity [2] can be written in the vicinal limit  $\phi \ll 1$  as

$$\Delta\rho g[\zeta(x) + \tau x] - \tilde{\alpha}(\phi) \frac{d^2\zeta(x)}{dx^2} = 0, \quad (1)$$

where  $\tilde{\alpha}(\phi) = \alpha + d^2\alpha/d\phi^2$  is the surface stiffness,  $\tau$  is the inclination angle of the facet, and  $\Delta\rho = \rho_s - \rho_l$  is the difference in densities of the solid and liquid phases of helium. Far enough from the edge, the first term in Eq. (1) dominates and the interface follows the direction of the gravitational horizon  $\zeta(x) = -\tau x$ , whereas closer to the facet border the ECS is explicitly controlled by the surface stiffness  $\tilde{\alpha}(\phi)$ .

Most of the present theories predict a decrease in the surface stiffness as  $\tilde{\alpha}(\phi) \propto \phi$  (see, e.g., Ref. [5]). The corresponding surface profile at  $\phi \rightarrow 0$  is expected to be  $\zeta(x) \propto x^{3/2}$  [12], which follows directly from Eq. (1). This is consistent with our data above  $\phi_{c2}$  but in striking contrast with the behavior seen at  $\phi < \phi_{c1}$ .

The angular dependence of the surface stiffness  $\tilde{\alpha}(\phi)$  at  $\phi \leq \phi_{c1}$  can be deduced from the measured profiles on the basis of Eq. (1). Over short intervals, where the first term in Eq. (1)  $E_g = \Delta\rho g[\zeta(x) + \tau x] \approx \text{const}$  with an accuracy of (20–30)%, an exponential solution is obtained if  $\tilde{\alpha}(\phi) = \kappa/\phi$ , where  $\kappa \approx x_0 E_g$ . [A more

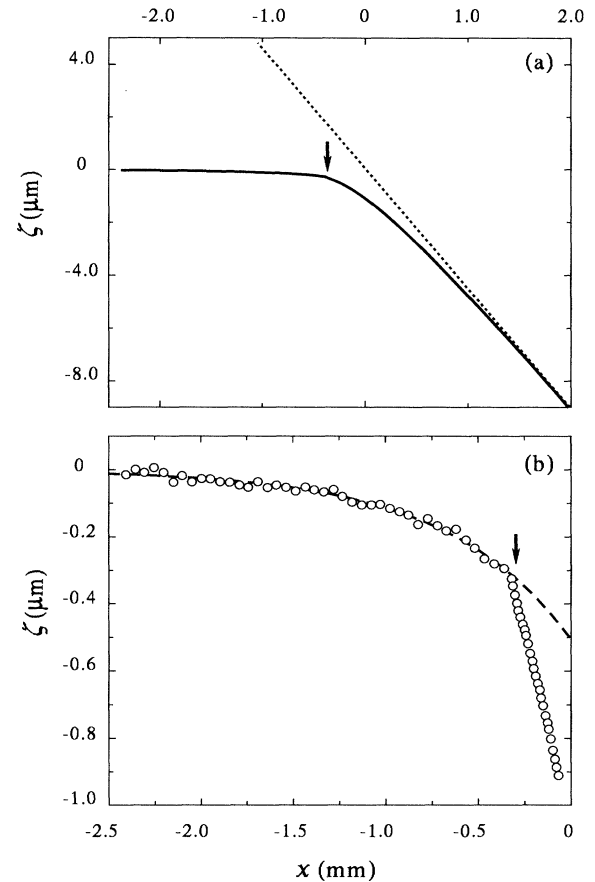


FIG. 2. Surface profile for one of our crystals with  $\tau = 4.5$  mrad, measured at 0.51 K. (a) Wide angular region:  $0 < \phi < \tau$ . Far enough from the edge, the interface follows the direction of the gravitational horizon  $\zeta(x) = -\tau x$  (dashed line), whereas closer to the facet edge the ECS is governed by the surface stiffness. (b) A magnified view of the same profile close to  $\phi = 0$ . The vertical arrow marks the point of the slope discontinuity. The solid curve corresponds to the best fit  $\zeta(x) = -\zeta_0 \exp(x/x_0)$ .

precise analysis can be done in the next approximation  $E_g \approx \Delta\rho g\tau x$ . In this case  $\tilde{\alpha}(\phi) = \kappa/\phi$  yields  $\zeta(x) \propto \text{erfc}(|x/\sqrt{2\kappa/\Delta\rho g\tau}|)$ , which in the range of our data is almost indistinguishable from an exponential function. However, the numerical analysis in this case is more complex and the results differ by less than 10% from the data presented in Fig. 3.] Results for four successive crystals with inclinations  $\tau = 5-15$  mrad are displayed in Fig. 3. As can be seen,  $\kappa$  varies approximately linearly as a function of temperature and we may summarize  $\kappa \approx (11 \pm 3) \times 10^{-4} T$  erg/cm<sup>2</sup> K.

The slope discontinuity can be considered as strong evidence of a first-order phase transition between two surface states [2,13]. Strong hysteretic behavior of crystal shapes is observed in our experiments. We often find

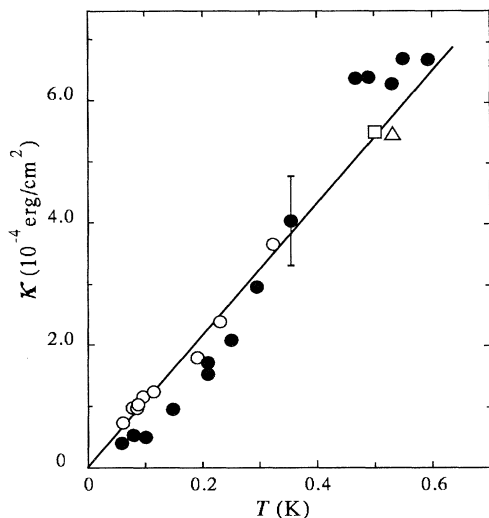


FIG. 3. Temperature dependence of the parameter  $\kappa$  for four samples:  $\tau = 4.5$  mrad ( $\square$ ),  $\tau = 5.3$  mrad ( $\triangle$ ),  $\tau = 14.9$  mrad ( $\circ$ ),  $\tau = 12.1$  mrad ( $\bullet$ ). The solid line corresponds to the best linear fit  $\kappa = (11 \pm 3) \times 10^{-4} T$  erg/cm<sup>2</sup> K.

jumplike changes in  $\phi_{c1}$ , caused by mechanical vibrations of the cryostat or by sudden temperature changes. For our data presented in Fig. 3,  $\phi_{c1}$  varies over the range  $50 \lesssim \phi_{c1} \lesssim 500 \mu\text{rad}$ .

Generally, a new surface state can be attributed to some special type of a spontaneous surface reorganization. A facet can be distorted due to the collective behavior of screw dislocations exposed on the surface. Frank-Read pairs of opposite screw dislocations are joined by sections of elementary steps on the interface. As shown by Uwaha and Nozières [14], polarization of such steps may result in a macroscopic tilt of the crystal boundary. The average concentration of Frank-Read sources can be estimated from the characteristic mobility threshold [5,14] for a uniformly growing facet:  $\Delta p = 2\beta/l$ , where  $\beta \approx 0.014$  erg/cm<sup>2</sup> [1,6], is the step energy and  $l$  is the average distance between neighboring dislocations. For our crystals,  $\Delta p$  is on the order of  $10^{-6}$  bar, which corresponds to  $l \approx 0.3$  mm, and yields at most an inclination angle  $\approx 10 \mu\text{rad}$  [14]. This estimation shows clearly that Frank-Read sources fail to explain the slope of the profile in Fig. 2(b). Moreover, it may be shown [11] that the dependence  $\tilde{\alpha} \propto 1/\phi$  cannot be explained on the basis of the model of Uwaha and Nozières with any fixed number of frozen-in dislocations.

The angular dependence  $\tilde{\alpha}(\phi) \propto 1/\phi$  can be obtained for several different reconstruction mechanisms. First, if in the new surface state the rotational symmetry  $C_3$  in the (0001) basal plane of an hcp helium crystal is broken, then the intrinsic surface stress tensor becomes anisotropic and, as a result, an elementary step now separates two domains having the same energy but different surface

stresses [15]. This difference, which depends strongly on the step orientation, gives rise to a net force on each step. In such a case, the elastic step-step interaction grows as  $\ln(d/a)$  with increasing interstep spacing  $d$  and, consequently,  $\tilde{\alpha}(\phi) \propto 1/\phi$ . However, at small angles  $\phi$  below some critical  $\phi_c^*$ , the interface becomes unstable against formation of elastic stress domains [15]. Hence, a doubly-reconstructed surface with ordinary  $1/d^2$  step-step interaction is formed at  $\phi < \phi_c^*$ , and the usual behavior of the surface stiffness,  $\tilde{\alpha}(\phi) \propto \phi$ , is regained [5,12]. Our experimental data at  $\phi < \phi_{c1}$  clearly contradict this scenario.

According to Andreev [16], flow of superfluid parallel to a facet results in the formation of steps oriented perpendicular to the velocity of the liquid  $v_s$ . Consequently, the stiffness of the reconstructed surface involves an additional term  $\tilde{\alpha}_v = \kappa_s/\phi = \rho a v_s^2/2\pi\phi$ . From the 50 nW heat leak to the experimental volume,  $v_s$  can be estimated to be at most on the order of  $10^{-8}$  cm/s, whereas  $v_s \approx 1 - 10$  m/sec would be needed to explain the experimentally observed values of  $\kappa$ .

Surface reconstruction can also be induced by thermal fluctuations of the crystal surface, which has been considered by Andreev [17]. These fluctuations correspond to microscopic Frank-Read sources and may be regarded as the surface part of thermally activated dislocation loops in the bulk [18]. According to Andreev, the presence of such loops results in a thermally excited plastic deformation of the entire crystal, which may destroy the crystal faceting. As discussed above, we have a similar situation in the case of usual quenched Frank-Read sources, distributed randomly on the crystal surface [14]. Assuming that Andreev's concept is valid, one may consider a simple statistical model that makes it possible to calculate the corresponding surface stiffness.

Each surface excitation, made of a microscopic Frank-Read source, generates a piece of elementary step of height  $a$  and length  $l$ , with some definite orientation of its normal vector  $\mathbf{n}$  in the plane of the facet. We assume here for simplicity that only two opposite orientations of these steps are possible: positive and negative. Consider a lattice gas of these excitations. The surface state is characterized by the average numbers of positive and negative steps  $N_+$  and  $N_-$  per area  $S$ . Both  $N_+$  and  $N_-$  must be much less than the total number of elementary cells  $N = S/s$ . If  $N_+$  is not equal to  $N_-$ , the gas of excitations becomes polarized, and an average tilt  $\phi$  of the surface appears:

$$\phi = S^{-1}(N_+ - N_-)al.$$

This situation differs from the case of quenched Frank-Read sources [14] because here the total number of excitations is not constant, and both  $N_+$  and  $N_-$  should be found by minimizing the surface free energy  $F$  at fixed

angle  $\phi$ :

$$F - N\lambda\phi = \min .$$

Here  $\lambda$  denotes the Lagrangian multiplier. For  $F$  we have the usual expression

$$F = \epsilon(N_+ + N_-) - T \ln \Gamma ,$$

where  $\epsilon$  is the activation energy, and  $\Gamma$  is the statistical weight of the surface state. After minimization we obtain

$$\phi = 2\chi e^{-\epsilon/T} \sinh(\lambda\chi/T) \quad (2)$$

and

$$F = -2NTe^{-\epsilon/T} \cosh(\lambda\chi/T) + N\lambda\phi , \quad (3)$$

where  $\chi = al/s$ . Equations (2) and (3) together give us the dependence  $F(T, \phi)$  at any  $T$  and  $\phi$ , provided that we can neglect interactions between excitations. We are interested in the case of relatively large angles  $\phi \gg \exp(-\epsilon/T)$  (but still  $\phi \ll 1$ ). Then we have from Eqs. (2) and (3)

$$F = \frac{N\phi}{\chi} \left[ \epsilon + T \left( \ln \frac{\phi}{\chi} - 1 \right) \right] ,$$

and finally for the surface stiffness

$$\tilde{\alpha}(\phi) = \frac{1}{S} \frac{\partial^2 F}{\partial \phi^2} = \frac{T}{\phi} (al)^{-1}. \quad (4)$$

We see that this mechanism yields correct angular and temperature dependencies of the surface stiffness; details of this model will be published elsewhere [11]. On the basis of Eq. (4) and our experimental data (Fig. 3), we estimate the effective length  $l \approx 30$  nm. For the activation energy  $\epsilon$ , we can only deduce a lower limit  $\epsilon \geq 10$  K.

Altogether, the shape of quantum crystals in our experiments has turned out to be more complicated than was expected on the basis of previous measurements [6]. The standard description [5] using interacting steps is not sufficient to explain the shape of our crystals in the vicinity of facets. Instead of a smooth transition from terraced planes to facets, we observe that crystal interfaces with  $\approx 2$  and  $\approx 0.2$  mrad inclinations are joined in a discontinuous manner. The  $1/\phi$ -dependent surface stiffness of the new state could be produced by dominant logarithmic interactions between steps separated by 1–100  $\mu\text{m}$ , but the reason for such forces is unclear. Our results can be explained by polarization of thermally induced dislocation loops terminating at the boundary. In this model, however, the required size of the loops is about 100 atomic spacings, which leads to questionably big excitation energies.

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\*Present address: P.L. Kapitza Institute for Physical Problems, ul. Kosygina 2, 117334 Moscow, Russia.

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