

# Anisotropy of the surface stiffness of He<sup>4</sup> crystals

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The angular dependence of the surface stiffness of hcp He<sup>4</sup> crystals has been measured.

The surface energy of a crystal,  $\alpha$ , and the surface stiffness  $\tilde{\alpha}_{ij} \equiv \alpha \delta_{ij} + (\partial^2 \alpha / \partial \theta_i \partial \theta_j)$  ( $i = 1, 2$ ;  $\theta_i$  are the angles which determine the crystallographic orientation of the surface), which is directly related to the surface energy, are extremely important characteristics, determining the equilibrium properties of a crystal surface. So far, however, there have been no direct measurements of the anisotropy of these two properties.

In this letter we report measurements of the angular dependence of the surface stiffness of He<sup>4</sup> crystals with an hcp structure. The surface stiffness is found by measuring the spectrum of crystallization waves.<sup>1</sup> The design of the apparatus, the procedure for growing oriented samples, the excitation method, and the method for detecting the crystallization waves are described in detail elsewhere.<sup>1</sup> The only substantial change was in the optical pressure chamber in which the crystals were grown and studied. In the present case the chamber was a cylinder (with an inside diameter of 23

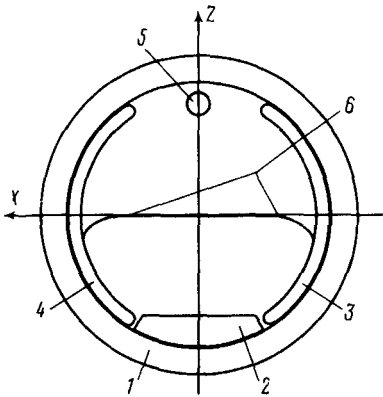


FIG. 1. The experimental chamber. 1—Housing; 2—bottom; 3, 4, 5—wire capacitors; 6—interface.

mm and length of 29 mm) whose symmetry axis ran horizontally and coincided with the optical axis of the cryostat and of the overall apparatus (Fig. 1). In the lower part of the chamber there was a flat bottom which was in thermal contact with a refrigerator. Furthermore, on the inside wall of the chamber, symmetrically on the left and right of the bottom, there were two wire capacitors, used to excite crystallization waves. To keep the experimental conditions symmetric with respect to a rotation of the chamber around its axis, we made both of these capacitors in the form of cylindrical surfaces bounded by a sector of  $120^\circ$ . Yet another wire capacitor (a cylinder 2 mm in diameter and 4 mm long), in the upper part of the chamber, was used as the source of a strong electric field during the growth of the oriented samples. The entire apparatus was mounted in the yoke of a ball bearing, which made it possible to rotate the sample, along with the chamber, through an angle of  $60^\circ$  in either direction from the horizontal position of the bottom.

The particular design of the apparatus makes it convenient to choose a Cartesian coordinate system whose  $Z$  axis runs vertically, whose  $Y$  axis is the rotation axis, and whose origin is at the center of the chamber. The  $X$  axis then becomes horizontal, running perpendicular to the rotation axis.

Under the experimental conditions, with the crystal filling the entire lower half of the chamber, the longitudinal dimension of the equilibrium interface was 29 mm, while the transverse dimension was 20 mm—substantially greater than the capillary constant, 1.4 mm. In this case, the maximum deviation from the horizontal for the parts of the surface at a distance of 5 mm or more from the chamber wall was  $\sim 3 \times 10^{-2}$  rad. A static voltage applied to the left and right capacitors significantly reduced the edge angle, with the result that the deviation of the surface from the horizontal in the  $XZ$  plane was reduced by an order of magnitude. This circumstance turned out to be particularly important in cases in which one of the crystallographic orientations corresponding to the atomically smooth state of the surface was represented in an equilibrium meniscus. We thus assume that the interface coincides with a horizontal plane at this accuracy level.

An alternating voltage of frequency  $\omega$  applied to one of the side capacitors excites

a plane crystallization wave with a wave vector  $\mathbf{k}$  directed parallel to the  $X$  axis. In this case the spectrum of crystallization waves  $\omega(\mathbf{k})$  can be described by the following expression, where we are ignoring a gravitational term:

$$\omega^2 = \frac{\rho_l |\mathbf{k}|}{(\rho_s - \rho_l)^2} \tilde{\alpha}_{ij} k_i k_j = \frac{\rho_l}{(\rho_s - \rho_l)^2} \tilde{\alpha} k^3,$$

where  $\rho_s$  and  $\rho_l$  are the densities of the solid and liquid phases, respectively,  $\tilde{\alpha} = \alpha + \partial^2 \alpha / \partial \theta^2$ , and the angle  $\theta$  is reckoned from the normal to the surface along the wave vector  $\mathbf{k}$ . Measurements of the spectrum for various orientations of the sample were carried out over the temperature interval 0.4–0.45 K, where the damping was quite low.

A study of the anisotropy of the surface stiffness is directly related to a study of roughening phase transitions. So far, three such phase transitions have been observed at the surface of hcp  $\text{He}^4$  crystals, with critical temperatures  $T_R$  of 1.28, 1.0, and 0.35 K for the crystallographic orientations (0001), (10 $\bar{1}$ 0), and (10 $\bar{1}$ 1), respectively.<sup>1–3</sup>

Using a technique developed previously,<sup>1,4</sup> we were able to grow crystals in which the angle between the  $C_6$  axis and the  $XZ$  plane was not more than a few milliradians. In the case of the two samples which we will discuss below, this angle was  $10^{-3}$  rad (sample No. 1) or  $2.5 \times 10^{-3}$  rad (No. 2). Ignoring this slight inclination, we assume that the  $C_6$  axis remains in the rotational plane in the course of the measurements, regardless of the rotation of the sample with respect to its original position. The orientation of the sample in the plane perpendicular to the  $C_6$  axis is arbitrary, and we were content with a crude estimate of it, within  $\sim 5^\circ$ . The angle  $\phi$ , between the  $C_2$  axis

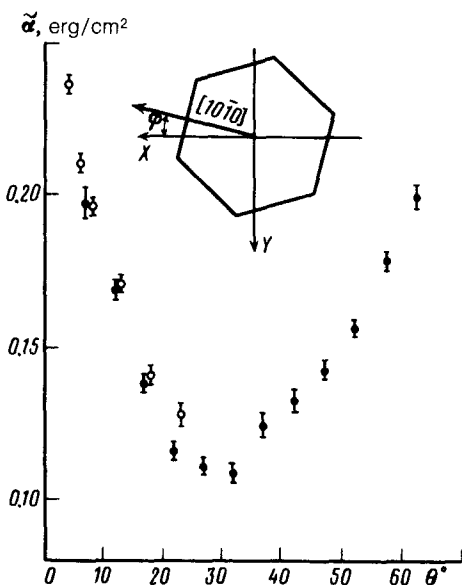


FIG. 2. Angular dependence of the surface stiffness of  $\text{He}^4$  crystals.  $\circ$ —Sample No. 1 ( $\phi = 5^\circ$ );  $\bullet$ —No. 2 ( $\phi = 13^\circ$ ).

of the crystal and the  $XZ$  plane (see the inset in Fig. 2), was  $5^\circ$  (No. 1) or  $13^\circ$  (No. 2) for the same two crystals.

When the chamber was rotated through an angle  $\theta$  in the course of an experiment, the interface assumed a horizontal position corresponding to the minimum energy in the gravitational field over a time on the order of a few seconds. This process occurred spontaneously, as a result of the melting of certain parts of the surface and the growth of other parts, while the orientation of the crystal with respect to the chamber was completely preserved. Each successive measurement of the spectrum thus corresponded to a crystallographic orientation rotated through an angle  $\theta$  with respect to the preceding measurement. This angle was measured within  $\sim 3 \times 10^{-3}$  rad by means of a telescope fitted with an angle gauge, which made it possible to measure the rotation in the vertical plane.

The method that we used has the serious limitation that only surfaces having an atomically rough structure have a growth coefficient large enough for the existence of weakly damped crystallization waves.<sup>1</sup> Near the crystallographic directions in an atomically smooth state, there is sharp increase in the damping due to the appearance of atomically smooth regions at the surface. In our case the minimum inclination angle of a vicinal face was  $4^\circ$  primarily because of the significant amplitude of the random vibrations of the surface caused by the vibrations of the cryostat. Figure 2 shows the functional dependence  $\tilde{\alpha}(\theta)$  for two samples, differing in the orientation of the  $C_2$  axis (the  $[10\bar{1}0]$  direction). The angle  $\theta = 0$  corresponds to the  $[0001]$  direction. Over the angular interval studied,  $4^\circ < \theta < 62^\circ$ ,  $\tilde{\alpha}$  was highly anisotropic, varying over the range 0.12–0.24 erg/cm<sup>2</sup>. Of particular interest is the increase in  $\tilde{\alpha}$  as  $\theta \rightarrow 0$ . This result contradicts the predictions of a new theory,<sup>5</sup> according to which  $\tilde{\alpha}$  should decrease linearly to zero as  $\theta \rightarrow 0$  at the temperatures and angles that we studied. In any case, this result means that the functional dependence  $\tilde{\alpha}(\theta)$  predicted by the theory can be manifested only at very small angles,  $\theta < 0.07$ . It is interesting to note that an extrapolation of our data to the value  $\theta = 0$  results in a good agreement with the results of a study<sup>4</sup> of the equilibrium shape of He<sup>4</sup> crystals obtained near the transition ( $T = 1.22$  K) of the (0001) plane. If this agreement is not simply fortuitous, we should state that  $\tilde{\alpha}$  has no significant temperature dependence.

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