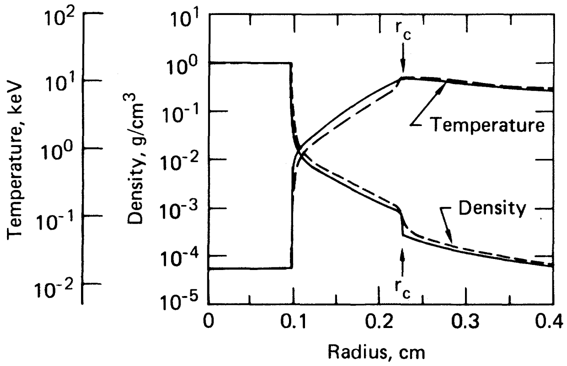

FIG. 3. Spatial profiles of density and temperature, for laser power $P_L = 78.5$ TW and other parameters as in Fig. 2. Solid lines show theoretical predictions; dashed lines are numerical results using the hydrodynamics code LASNEX. The two agree quite well.

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Interface between Superfluid and Solid ^{4}He

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With an optical technique it is found that the equilibrium interface between superfluid and hcp ^{4}He is partially faceted, showing that at least some of it is atomically smooth. This conclusion is also consistent with the behavior of the crystal during melting and growth. The surface tension α_{LS} on the rounded part of the interface is found to be independent of temperature.

PACS numbers: 67.40.Kh, 67.80.Gb, 68.45.–v

Andreev and Parshin (AP) have discussed[1] the theory of the ^{4}He crystal-superfluid interface. They conjecture that the interface is atomically rough, even at $T = 0$, due to nonlocalized, zero-point quantum-mechanical defects. Since the surface is rough they predict the interfacial surface tension α_{LS} to be a smooth function of the surface orientation with respect to the crystal axes. It follows that the equilibrium shape of the crystal should be rounded, with no facets, in agreement with several experimental observations.[2] AP also predicted the existence of "melting-freezing" capillary waves on the interface. These have recently been discovered by Keshishev, Parshin, and Babkin.[3]

This Letter reports results of a study with use of an optical-holographic technique.[4] In general, our observations confirm the fluidlike behavior of the interface; however, we do observe stable equilibrium facets on the hcp crystal. The latter observation leads us to conclude at least part of the interface to be atomically smooth. By studying the growth and melting of the hcp crystals we find that the growth coefficient is anisotropic. The anisotropy is consistent with part of the surface being atomically smooth and part atomically rough.

In a recent Letter Balibar, Edwards, and Laroche[5] (BEL) reported the first direct measurements of α_{LS} between superfluid ^{4}He and bcc and hcp ^{4}He crystals, and also the angle of contact θ between the interface and a copper surface. Their results were consistent with the crystals' having a shape similar to that of a nonwetting liquid —i.e., a gravitationally determined convex meniscus. In addition BEL found a strong temperature dependence of α_{LS} and θ below 1 K. The latter was unexpected since it implied the surface entropy $-d\alpha_{LS}/dT$ to be large near 1 K but small both above and below this temperature.

From measurements of the unfaceted part of the meniscus profile we have determined the interfacial tension and contact angle. We find no temperature dependence to either α_{LS} or θ to within our experimental error, in distinction to BEL. However, our values for the capillary rise, $\alpha_{LS}\cos\theta$, are in good agreement with BEL.

Our experimental procedure is extremely simple. We grow the crystal between the parallel glass windows of our optical cell and then photograph the liquid-solid interface. The cell shown schematically in the inset of Fig. 1, contains a disklike sample of He that is 3.6 mm thick and has an observable diameter of 9 mm. The symmetry axis of the disk lies in the horizontal plane and is parallel to the axis of the optical system as is shown schematically in Fig. 3 of Ref. 4. The optical system allows us to use standard microscopic techniques, real-time interferometric holography or shearing interferometry.

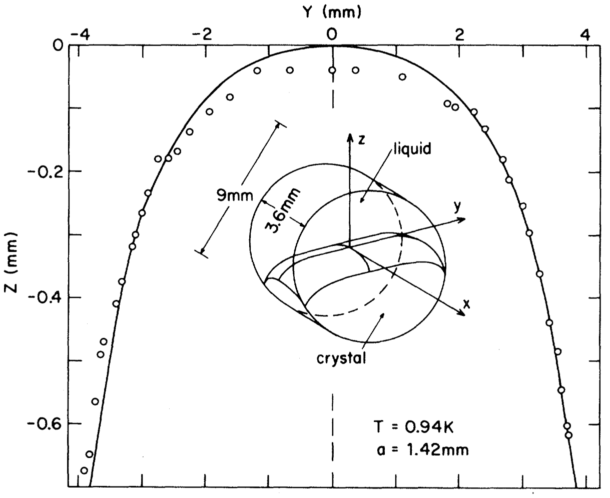

FIG. 1. Measured meniscus (circles) compared with a calculated profile, at $x = 0$. Inset, a perspective sketch showing the cell geometry and the liquid-solid interace.

We have usually been able to nucleate a single crystal of either bcc or hcp ^{4}He, compressing the superfluid phase by admitting gas from a room-temperature supply via a filling capillary. Single crystals do not grow from the normal fluid phase. The hcp crystal tends to nucleate from one of several isolated nucleation sites near the copper wall or indium seals of the cell. During growth at temperatures below 1 K, the hcp crystals are always completely faceted with angular interfaces when the growth rate exceeds about 1 mm/sec [see Fig. 2(a)]. We can measure the angles between the facets from the interferograms and thus determine the crystal orientation. All the facets we have observed correspond to principal axes.

During rapid melting the hcp crystals are always completely rounded. This distinction between growth and melting is well known for classi-

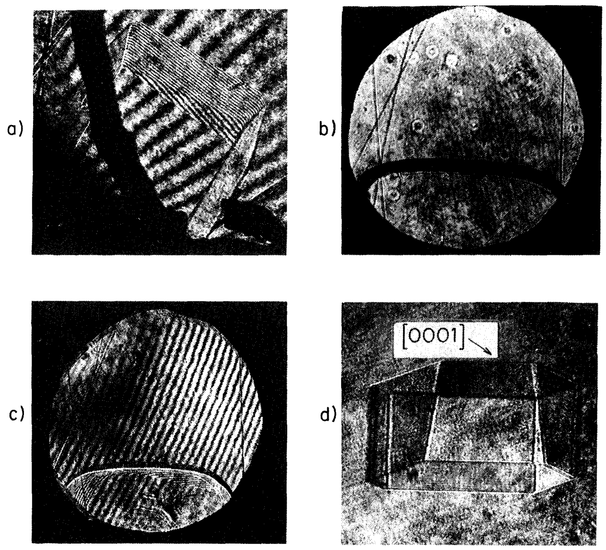

FIG. 2. (a) A faceted hcp crystal ($T = 0.55$ K) during fast growth (holographic interferogram). (b) An equilibrium gravitational hcp meniscus at $T = 0.6$ K (spatially filtered image). (c) Stable facets on a meniscus at $T = 0.77$ K (holographic interferogram). (d) A stable hcp crystal, about 2 mm high, attached to one window and grown from a 0.1% ^{3}He mixture at $T = 0.4$ K (interference image).

cal crystals[6] and is due to an anisotropic melting/growth coefficient, which has cusps at the principal faces. During growth, the principal faces limit the extent and shape of the crystal, whereas during melting the nonprincipal faces consume the principal faces.

An interesting observation is that after melting the hcp crystal by reducing the pressure by as much as one atmosphere below the melting curve, recompressing results in a crystal having an identical orientation; if the pressure drop is larger, the new crystal forms with an unrelated orientation. Memory of the orientation is most likely due to a layer of solid He on the cell wall, stabilized near the substrate by the van der Walls attraction.[7]

In contrast to hcp, the bcc crystal usually nucleates simultaneously at a number of sites. Even more important, bcc crystals are always rounded during growth, in equilibrium and while melting.

Once growth has ceased, a large hcp crystal seems to undergo a wobbly sort of flow until the interface has been transformed from its angular form to a gravitationally determined meniscus [Fig. 2(b)]. Figure 1 shows that this meniscus can be compatible with the profile predicted by the Laplace-Young equation for the meniscus between two liquids

$$\Delta P=(z_0-z)(\Delta\rho)g=\alpha_{LS}(1/R_1-1/R_2), \qquad (1)$$

where ΔP and $\Delta\rho$ are the pressure and density differences across a curved interface and R_1 and R_2 are the principal radii of curvature, and $\Delta P=0$ at z_0. There are notable exceptions to this result. Firstly, below 1 K if the crystal dimensions are sufficiently small we have often observed stable hexagonal structures with rounded edges showing no tendency of transformation to a completely rounded meniscus. Their maximum size is of the order of the capillary constant $a=[2\alpha_{LS}/(\Delta\rho)g]^{1/2}$ which is 1.4 mm for the hcp surface. Secondly, even large crystals with mostly rounded surfaces, display small facets [Fig. 2(c)]. These are particularly large when a principal plane is close to horizontal. In general, the vertical extent of a stable facet is not much larger than a. The stable facets seem to represent a true equilibrium state since they resist all attempts at removal by melting to miniscule dimensions and subsequent regrowth. By contrast, bcc menisci are never faceted, although they sometimes show asymmetric features.

The fact that hcp crystals are partially faceted indicates, from Wulff's theorem,[6] that the polar diagram of the angular dependence of α_{LS} possesses cusps in some principal directions. This is in partial contradiction with the assertion of Andreev and Parshin that *all* of the interface is atomically rough, even at 0 K. The faces corresponding to cusps must be atomically smooth. On the other hand, the anisotropy of the melting/growth coefficient, mentioned above, is consistent with the rounded part of the crystal surface being atomically rough. At least for classical crystals, atomically rough surfaces have much larger growth coefficients than atomically smooth faces.

The determination of α_{LS} and θ from the observations requires a solution of (1) for the geometry of the cell. Moreover, if α_{LS} is anisotropic, α_{LS}/R_i in (1) should be replaced by $(\alpha_{LS}+\partial^2\alpha_{LS}/\partial\varphi_i^2)/R_i$, where φ_i is the angular variable in the plane of R_i.[8] An appropriate three-dimensional solution of (1) is difficult to find, and so we have assumed that the x and y dependencies can be decoupled to a good approximation. The surface tension is found by comparing the measured profile of the meniscus in the y-z plane with the two-dimensional solution of (1) which is calculated by a Simpson's-rule integration following the formulation of Buff.[9] The parameter a is varied with the aim of obtaining the best visual fit between the calculation and the data, both displayed on the screen of a minicomputer. Then the value of $\alpha_{LS}\cos\theta$ is constrained to agree with a measurement of the height of the meniscus in the x-z plane at the center of the cell. The photographed profiles are measured to an accuracy of 0.03 mm and the error bars on the results (Fig. 3) indicate the limits of reasonable visual fit. When stable facets were present, only the curved part of the meniscus was fitted.

Our results for α_{LS} and that of Keshishev, Parshin, and Babkin[3] disagree with those of BEL (also shown in Fig. 3) by as much as a factor of 4 at low temperatures, although above 1 K they agree very well. At all temperatures the values of $\alpha_{LS}\cos\theta$ agree within experimental error. The disagreement in α_{LS} leads to different results for θ. In our experiment $\alpha_{LS}=0.16\pm0.03$ erg cm^{-2} and $\theta=137°\pm10°$ for hcp, while for bcc we find $\alpha_{LS}=0.084\pm0.015$ erg cm^{-2} and $\theta=146°\pm10°$, both independent of temperature. Our photographs clearly confirm the qualitative conclusion of BEL that solid ^{4}He does not wet copper (and in our case glass also) as well as the superfluid does.

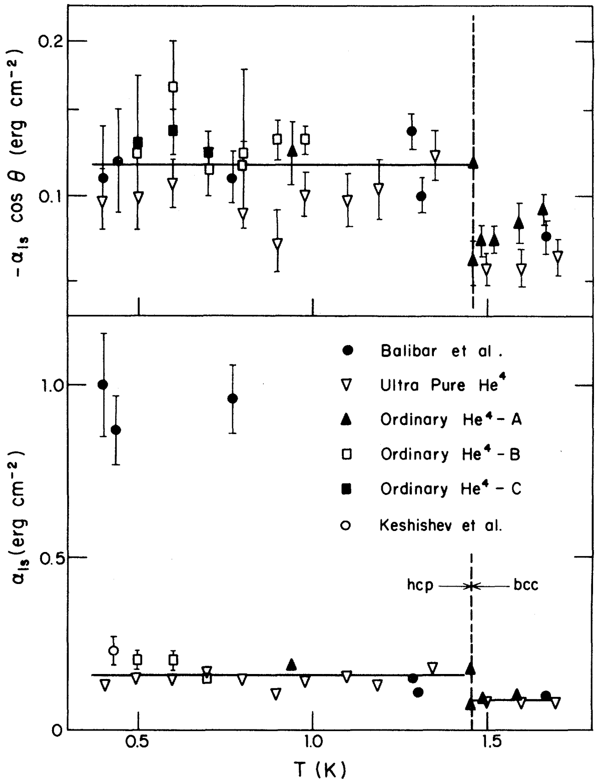

FIG. 3. Summary of experimental results. Above: the capillary rise $\alpha_{LS}\cos\theta$, below: α_{LS} as a function of temperature. *A*, *B*, and *C* represent three independent experimental runs with different crystal orientations.

The reason for the discrepancy may be as follows. The present observations show that, in the BEL experiments the "bubble" of solid used in the determination of α_{LS} was certainly faceted below 1 K. Their experiment therefore measured an angular average of $\alpha_{LS}+\alpha_{LS}''$ where $\alpha_{LS}'' = \partial^2\alpha/\partial\varphi^2$. Our experiment also measures an average of $\alpha_{LS}+\alpha_{LS}''$ over the curved part of the meniscus only. The existence of facets implies that α_{LS} has minima in principal directions, so that α_{LS} is less for a facet than for a curved surface. The key to the puzzle is the α_{LS}'' term. For example, a two-dimensional hexagonal model having a surface-tension contribution proportional to the density of broken nearest-neighbor bonds (see Ref. 8),

$$\alpha + \alpha_0 \cos[(\varphi \bmod 60°) - 30°] \tag{2}$$

leads to

$$\alpha + \alpha'' = \alpha_0 \delta(\varphi - n60°). \tag{3}$$

The surface tension α is cusped and gives six facets, whereas $\alpha+\alpha''$ is unchanged *except* at the facets. The change would enhance the measured value of α in the BEL type of experiment. It is then necessary to assume that the cusped part of α_{LS}, and the facets, disappear above 1 K. This is consistent with the present observations.

A factor which cannot be completely ignored is the possible influence of ^{3}He impurity. We find that ^{3}He fractions exceeding 10^{-4} enhance the stable facets and eliminate the rounded parts of the surface [see Fig. 2(d)]. One possibility considered was that ^{3}He was preferentially adsorbed at the interface between the liquid and the solid, analogous to the situation at the liquid-vapor interface.[10] Nominally pure ^{4}He well gas contains 0.1 ppm ^{3}He. This would be more than enough to complete a monolayer at the interface. We can now rule out this possibility as a result of our repeating the experiment with ultrapure ^{4}He, obtained by filtering out the ^{3}He with a porous Vycor glass superleak. Previous experience[11] has shown that such ultrapure ^{4}He contains no more than a 10^{-9} ^{3}He fraction. No significantly different behavior was observed.

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High-Resolution Heat-Capacity Studies near the Nematic-Smectic-*A* Transition in Octyloxycyanobiphenyl (8OCB)

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High-resolution ac calorimetry measurements on two different samples of 8OCB whose transition temperatures differ by less than 0.1 K show different critical behavior at the nematic–smectic-*A* transition. $\alpha^+ = \alpha^-$ is near zero on one sample, agreeing with Johnson's measurements, while $\alpha^+ = \alpha^-$ equals 0.25 on the second sample which agrees with Garland's measurements on 8OCB. These results suggest the possibility that the nematic–smectic-*A* transition on 8OCB is near a tricritical point or crossover between two types of critical behavior.

PACS numbers: 64.70.Ew

In recent years there has been much theoretical and experimental work aimed toward the understanding of the nematic–smectic-*A* phase transition. The mean-field theories of McMillan[1] and Kobayashi[2] describe the smectic-*A* order parameter as a complex scalar, $\psi = |\psi| e^{i\varphi}$, where $|\psi|^2$ is the amplitude of the smectic layers and $\varphi = 2\pi z/d$, where d is the interlayer spacing. McMillan has predicted that the nematic–smectic-*A* transition can be second order and that the tricritical point, where the transition changes from second to first order, would occur at some critical value of the ratio of the nematic–smectic-*A* transition temperature to the nematic-isotropic transition temperature, T_{na}/T_{ni}. Since then there has been extensive work on pure compound which exhibit this transition in the effort to determine the order of the transition and to measure the critical exponents when it is a second- or slightly first-order transition. de Gennes[3] has predicted heliumlike exponents based on the expression for the Landau free energy of the smectic-*A* phase. Several experiments have been done to measure the exponents and compare them to the helium values of $\nu = 0.66$ and $\alpha = 0$. Johnson *et al.* have measured α in pentylphenylthiol-octyloxybenzoate ($\bar{8}$S5)[4] and in 8OCB[5] and have found $\alpha^+ = \alpha^-$ near zero. In their 8OCB measurements they observe a 40-mK-wide flattened region at the transition. Birgeneau, Litster, and co-workers[6] have done light-scattering and x-ray-scattering measurements and have found perpendicular and parallel correlation lengths which diverge at different rates. In the case of different $\nu_\parallel$ and $\nu_\perp$, an anisotropic scaling law can be written

$$2\nu_\perp + \nu_\parallel = 2 - \alpha.$$

Using anisotropic scaling with results of Ref. 6 on 8OCB, $\nu_\parallel = 0.71$, $\nu_\perp = 0.58$, we obtain $\alpha = +0.12$. In another ac heat-capacity experiment on 8OCB, Garland *et al.*[7] has measured α between 0.2 and 0.3 which does not agree with the value of Ref. 5 or with the value which emerges from Ref. 6. The sample used by Garland *et al.* is from the newer of two batches made by British Drug Houses (BDH).

We chose 8OCB for measurement because of its chemical stability. We decided to do measurements on samples from the two different batches of 8OCB because there existed such varying results for the exponent α found by the other groups. The samples of 8OCB were given to us by P. E. Cladis of Bell Laboratories and D. Johnson of Kent State University. Both samples originated from BKH but were reportedly from different stock. Cladis had vacuum sublimated the 8OCB from the earlier stock, while Johnson's sample

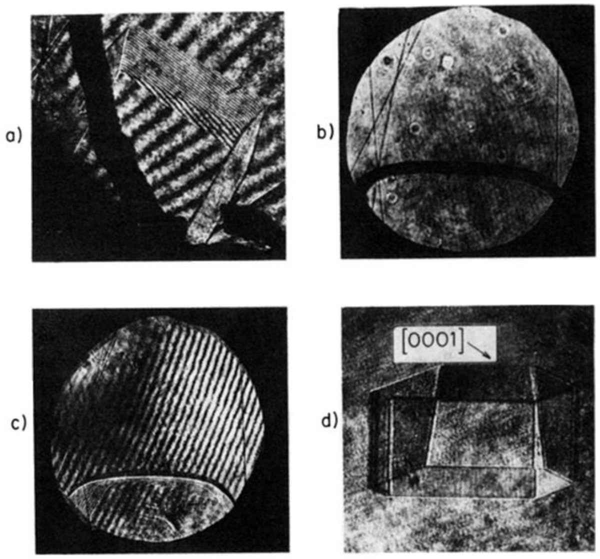

FIG. 2. (a) A faceted hcp crystal ($T = 0.55$ K) during fast growth (holographic interferogram). (b) An equilibrium gravitational hcp meniscus at $T = 0.6$ K (spatially filtered image). (c) Stable facets on a meniscus at $T = 0.77$ K (holographic interferogram). (d) A stable hcp crystal, about 2 mm high, attached to one window and grown from a 0.1% ^{3}He mixture at $T = 0.4$ K (interference image).