

Identification of the facets of ^4He crystals

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The crystallographic orientation of the faceting of ^4He crystals has been determined. The $[1011]$ roughening transition has been studied.

Roughening transitions corresponding to three crystallographically nonequivalent directions have been observed in ^4He at temperatures of 128 K (Refs. 1 and 2), and 0.365 K (Ref. 3). A comparison of the symmetry of the hcp structure of ^4He and the observed morphological features of the crystals makes it possible to identify the facet perpendicular to the C_6 axis, with a transition temperature $T_{R1} = 1.28$ K, as the (0001) facet. It also follows from symmetry considerations that in the case of the lateral facet of a prism ($T_{R2} = 1$ K) a choice must be made between $(10\bar{1}0)$ and $(11\bar{2}0)$ planes. With regard to the third phase, it has been learned that the vector normal to it lies in a plane which passes through the normal to the first two facets. In addition, the third facet makes an angle of $60^\circ \pm 3^\circ$ with the (0001) plane.³ These two conditions are satisfied by either the $(10\bar{1}1)$ or $(11\bar{2}2)$ plane, depending on the orientation of the lateral facet.

This ambiguity has been mentioned in one way or another in all previous publications on the roughening transitions in ^4He . In a paper by a Parisian group,⁴ however, a decision was made in favor of the $(10\bar{1}0)$ and $(10\bar{1}1)$ facets. The condition on which that decision was based is the possibility of satisfying the known universal relation

$$kT_R = \frac{2}{\pi} (\det \tilde{\alpha}_{ij})^{1/2} d^2, \quad (1)$$

which determines, for a given crystallographic orientation, the relationship among the transition temperature T_R , the surface stiffness $\tilde{\alpha}_{ij}$ ($T = T_R$), and the distance between plane d . [We recall that, in general, we would have $\tilde{\alpha}_{ij} \equiv \alpha \delta_{ij} + \partial^2 \alpha / \partial \theta_i \partial \theta_j$ ($i = 1, 2$),

where α is the surface energy, which depends on the crystallographic orientation θ_i .]

The $[0001]$ and $[10\bar{1}1]$ directions obviously make an angle $\arctan \frac{1}{3}\sqrt{2} \approx 62.1^\circ$, while the $[0001]$ and $[11\bar{2}2]$ directions make an angle $\arctan 2\sqrt{\frac{2}{3}} \approx 58.5^\circ$. It thus becomes possible to directly determine the crystallographic orientation of the facets.

This letter is, in all regards, a continuation of research begun previously. For an explanation of all the necessary details, we refer the reader to two previous papers.^{5,6} By chance, one of the test samples turned out to be oriented in such a way that the deviations of the normals \mathbf{n}_1 (basis), \mathbf{n}_2 (lateral facet), and \mathbf{n}_3 (oblique facet) from the XZ plane, which is perpendicular to the chamber rotation axis (Fig. 1; Ref. 5), were $+6 \times 10^{-3}$ rad, -5×10^{-3} rad, and -2×10^{-3} rad, respectively. The angle between the first and third facets was measured, within the deviation of the vectors \mathbf{n}_1

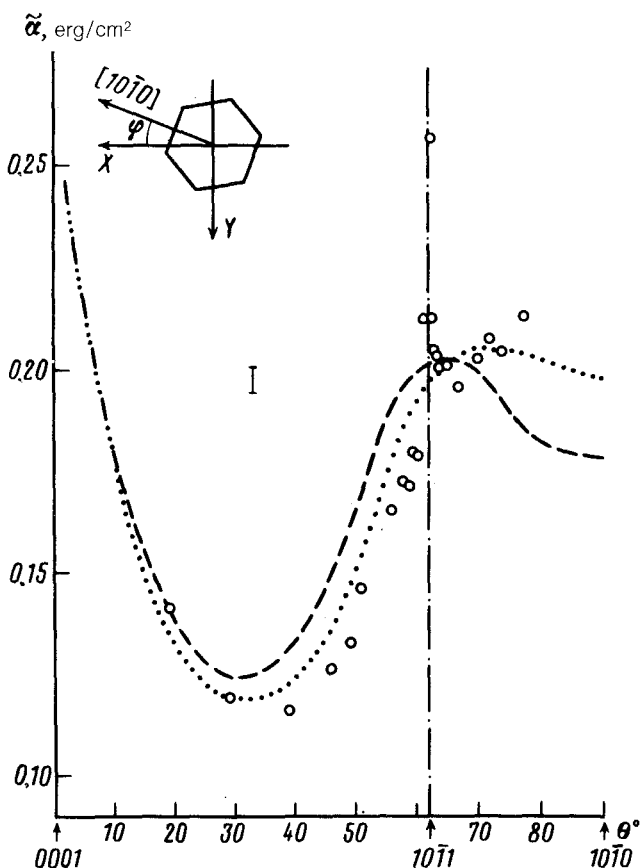


FIG. 1. Angular distribution of the surface stiffness in three cross sections passing through the C_6 axis. Here θ is the angle between the normal to the surface and the $[0001]$ direction, and φ is the angle between the intersection of the XZ cross section and the $[10\bar{1}0]$ direction (see the inset). ●— $\varphi = 0$; dotted line— $\varphi = 10^\circ$; dashed line— $\varphi = 30^\circ$. The arrow below the abscissa showing the $[10\bar{1}1]$ direction refers only to the $\varphi = 0$ cross section.

and n_3 from the frontal plane (the associated relative error was not greater than 0.02%), on a slowly growing crystal with the help of a telescopic tube fitted with a goniometer (the angular resolution was 1'). The result of this measurement was $62.0^\circ \pm 0.2^\circ$. The oblique facet is thus identified unambiguously as the $(10\bar{1}1)$ facet; the lateral facet is then identified as $(10\bar{1}0)$.

Using this unique sample again, we studied (as in Refs. 5 and 6) the anisotropy of the surface stiffness $\bar{\alpha}$ and the growth coefficient K in the cross section perpendicular to the $[11\bar{2}0]$ direction. Figure 1 shows the measurements of the surface stiffness in this cross section at a temperature of 0.33 K. The angle θ is reckoned from the C_6 axis; the $[10\bar{1}1]$ direction, which is the direction of greatest interest, corresponds to the value $\theta = 62.1^\circ$. Also shown here are smoothed angular distributions $\bar{\alpha}(\theta)$ for two other cross sections passing through the C_6 axis, one of which (the dashed line) coincides within $\sim 1^\circ$ with the $(10\bar{1}0)$ plane, while the other (dotted line) makes an angle $\varphi \approx 10^\circ$ with the $[10\bar{1}0]$ direction. These results were found in previous experiments at a temperature of 0.4 K. The error bar at the middle of this figure corresponds

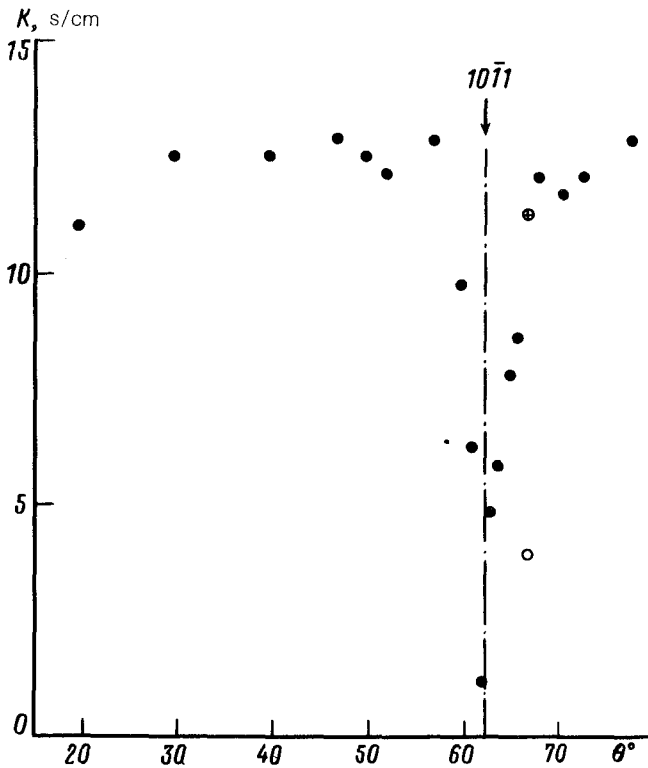


FIG. 2. Angular distribution of the growth coefficient in the $(11\bar{2}0)$ cross section ($\varphi = 0$). ●— $T = 0.33$; ○— $T = 0.43$ K; ⊕— $K(T = 0.43 \text{ K}) \times 0.43/0.33$ (Ref. 4).

to the mean square deviation of the experimental points from the average curves. Since there is no significant azimuthal anisotropy near the [0001] direction, we restricted our study of the $(11\bar{2}0)$ cross section at small angles to the value $\theta = 19.3^\circ$. Unfortunately, the sample was originally oriented in such a way that even a maximum possible rotation of the chamber brought us no closer than 12.6° to the $[10\bar{1}0]$ direction. In addition to the significant anisotropy of the stiffness as a whole [which is extremely similar to the behavior of $\tilde{\alpha}(\theta)$ in other cross sections passing through the C_6 axis], we see a structural feature at $\theta = 62^\circ$ in Fig. 1. The sharp increase in $\tilde{\alpha}$ as both sides of the $(10\bar{1}1)$ facet are approached is, according to previous measurements,⁵⁻⁷ a characteristic feature of an atomically smooth state of this facet. Equally typical is the vanishing of the growth coefficient K (Fig. 2), which is due to an increase in the damping of crystallization waves. We find only a quantitative difference from the results for the (0001) and $(10\bar{1}0)$ facets:⁶ The anomalous behavior of $\tilde{\alpha}(\theta)$ and $K(\theta)$ near the $(10\bar{1}1)$ facet is limited to a much narrower angular interval, about $\pm 5^\circ$.

Under the assumption that the temperature of the third transition is³ $T_{R_3} = 0.365$ K, we attempted to follow the changes in the structural features in $\tilde{\alpha}(\theta)$ and $K(\theta)$ caused by an increase in the temperature and by the transition of the $(10\bar{1}1)$ facet to an atomically rough state. We assumed that the growth coefficient, being strictly zero on an atomically smooth facet, should increase sharply at the transition, reaching a value characteristic of a rough surface at the given temperature. The need for this sort of disappearance of the $K(\theta)$ structural feature is quite obvious, and it is supported by measurements of the temperature dependence of the mobility of the (0001) facet near T_{R_1} (Ref. 4). We measured the surface stiffness and the growth coefficient over the temperature interval 0.33–0.43 K for two orientations of the surface: $\theta = 62.4^\circ$ and $\theta = 66.4^\circ$. In contrast with expectations, we observed no qualitative changes in the angular distribution $\tilde{\alpha}(\theta)$ or $K(\theta)$. Because of the strong damping in the orientation $\theta = 62.4^\circ$, it was not possible to measure the stiffness or the growth coefficient when the temperature was raised, even if only slightly. With regard to the second orientation, $\theta = 66.4^\circ$, we should argue that there are no changes in $\tilde{\alpha}$ —no changes significant at our error level—over the interval to a temperature of 0.43 K. The observed decrease in the growth coefficient, on the other hand, can be explained in a trivial way, by means of the known temperature dependence² $K \propto T^{-4}$. The latter circumstance is illustrated by two points in Fig. 2; the lower of the two is the result of a measurement at $T = 0.43$ K, while the upper corresponds to this value of K , normalized to the temperature dependence $K(T)$. Unfortunately, we were not able to carry out measurements at a higher temperature.

These experimental data show that the atomically smooth state of the $(10\bar{1}1)$ surface persists to a temperature of at least 0.43 K and that the condition $T_{R_3} > 0.43$ K holds. It is also clear that the value $T_{R_3} > 0.365$ K, which arose in an interpretation of indirect experiments,³ is considerably too low.

Extensive theoretical work, based in part on exactly solvable models and a renormalization-group analysis, leads to several universal results, which directly establish the relationship between roughening transitions and a Kosterlitz-Thouless transition. One of these results is relation (1), which has, in the opinion of several authors,^{4,8,9} received convincing experimental confirmation. A linear angular dependence $\tilde{\alpha}(\theta)$

near singular directions also follows from a renormalization-group study which has been worked out in detail.^{4,10} The monotonic increase in $\bar{\alpha}(\theta)$ toward the [0001] direction (Fig. 1) and the [10 $\bar{1}$ 0] direction (Fig. 1; Ref. 6), up to an angle of 3×10^{-2} at $T = 0.4$ K (which is quite low in comparison with T_{R1} and T_{R2}), is itself a fairly weighty counterargument. Since there is no significant temperature dependence of the surface stiffness either above or below T_R (according to the results presented here and numerous previous studies^{2,4,6,7}), this assertion applies equally well to the [10 $\bar{1}$ 1] orientation (Fig. 1), except that in this case the minimum experimental angle is 6×10^{-3} rad. The suggestion that a dependence $\bar{\alpha} \propto \theta$ should persist at even smaller angles raises serious objections, since in this case the ratio d/ξ (ξ is the correlation length), which determines the region of critical angles, turns out to be anomalously small even at $T \ll T_R$. Nevertheless, we will adopt that assumption, and we will use the theory which we have been discussing to examine the experimental data.

Figure 3 shows smoothed angular curves of the diagonal components of the tensor $\tilde{\alpha}_{ij}$. The orientation of the normal to the surface is determined by the polar angle θ and by the longitude φ in the polar coordinate system of the crystal lattice (the inset in Fig. 3). Although it is not complete, this diagram does demonstrate the general picture of the anisotropy of the surface stiffness. Near the [0001] direction, with $\theta = 5 \times 10^{-3}$ rad, we have $\tilde{\alpha}_{11} = \tilde{\alpha}_{22}$, and the value 0.33–0.36 erg/cm² is reached.⁷ Under the assumption made above, this value should evidently be thought of as a lower limit with respect to $\tilde{\alpha}_{ij}$ ($\theta = 0, T = T_R$). This comment also applies to two other singular orientations. Furthermore, the molar volume of ¹¹ 20.983 cm³/mole corresponds to a distance between planes $d_{0001} = 2.99$ Å. From (1) we find the value $T_{R1} = 1.36$ K.

Near the [10 $\bar{1}$ 0] direction, two principal values of the stiffness tensor have been measured: $\tilde{\alpha}_{22}(\theta = 99^\circ, \varphi = 1.5^\circ) = 0.36$ erg/cm² (Ref. 6) and $\tilde{\alpha}_{11}(\theta = 77.4^\circ, \varphi = 0) = 0.21$ erg/cm² (Fig. 1). Because of the particular features of $\tilde{\alpha}_{11}$ and $\tilde{\alpha}_{22}$ in the immediate vicinity of each of the atomically smooth facets, it seems completely natural to extrapolate the dependence $\tilde{\alpha}_{11}(\theta, \varphi = 0)$. This extrapolation, shown by the dashed lines in Fig. 3, leads to the value $\tilde{\alpha}_{11}(\theta = 88^\circ, \varphi = 0) \approx 0.25$ erg/cm². The translational period along the [10 $\bar{1}$ 0] direction is $d_{10\bar{1}0} = 3.18$ Å. According to (1), the transition temperature is $T_{R2} = 1.40$ K (the experimental value is 0.9–1.05 K).

Near the [10 $\bar{1}$ 1] direction, only one of the principal values has been measured: $\tilde{\alpha}_{11}(\theta = 62.1 \pm 0.3^\circ, \varphi = 0) = 0.22$ erg/cm² (Fig. 1). We can use the estimate $\tilde{\alpha}_{22}(\theta = 62.1^\circ, \varphi = 0) \approx 0.2$ erg/cm², which looks to us to be totally realistic. The translational period $d_{10\bar{1}1}$ is 2.80 Å. The theoretical value found as a result, $T_{R3} \approx 0.76$ K, looks as pessimistic as that in the preceding case, even in light of the new experimental data.

We believe that this picture completely refutes the idea that there is a quantitative or even qualitative agreement on this question.

We note in conclusion that for the [11 $\bar{2}$ 0] ($d_{11\bar{2}0} = 1.83$ Å) orientation both principal values of the stiffness in the rough state have been measured directly. The results are $\tilde{\alpha}_{11} = 0.18$ erg/cm² and $\tilde{\alpha}_{22} = 0.08$ erg/cm². In this case, expression (1) yields a transition temperature of 0.18 K. Research results which can be interpreted

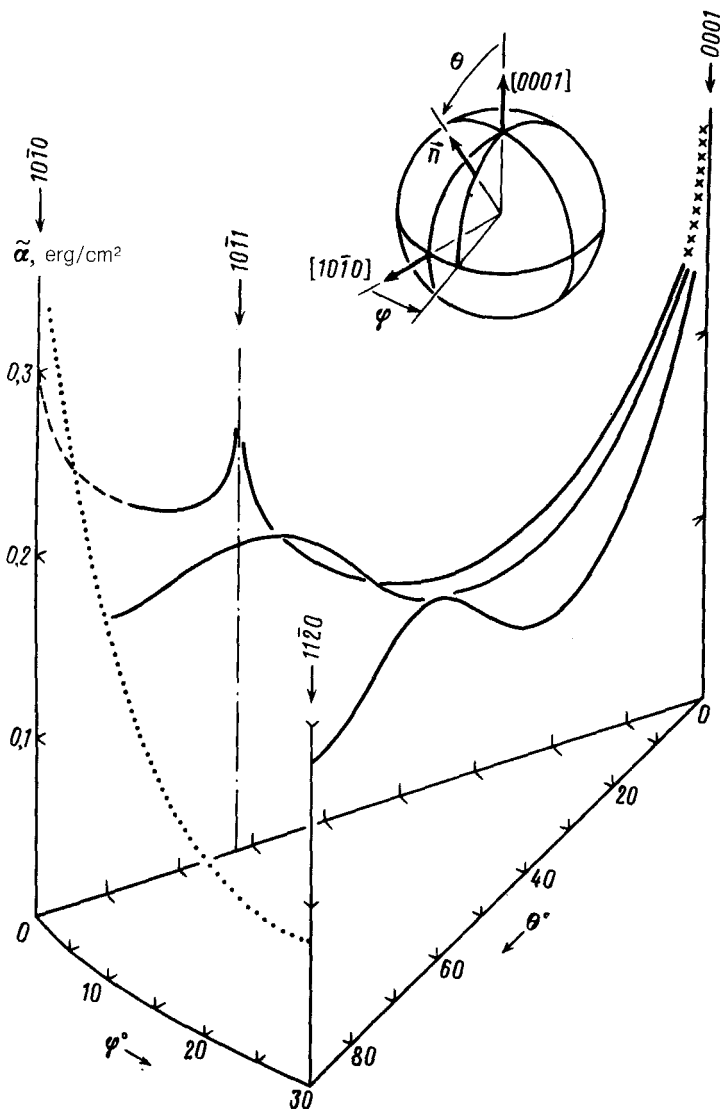


FIG. 3. Anisotropy of the diagonal components of the surface-stiffness tensor. Solid lines— $\bar{\alpha}_{11}(\theta, \varphi = \text{const})$ in the cross sections $\varphi = 0$, $\varphi = 10^\circ$, and $\varphi = 30^\circ$ (according to the results of the present study); dashed line—extrapolation of $\bar{\alpha}_{11}(\varphi = 0, \theta \rightarrow 90^\circ)$; \times — $\bar{\alpha}_{11}(\theta, \varphi = \text{const})$ at small angles θ and at $T = 1.22$ K (Ref. 7); dotted line— $\bar{\alpha}_{22}(\varphi, \theta = 90^\circ)$ (Ref. 6).

unambiguously in the corresponding angular and temperature regions will make it possible to finally resolve this problem.

COMMENT REGARDING A PREVIOUS PUBLICATION⁶

We apologize to the readers and the editors for a calculation error which had the result that the values of the growth coefficient K , shown in Fig. 2 of Ref. 6, were too low by a factor of 2.

- ¹J. E. Avron, L. S. Balfour, C. G. Kuper *et al.*, Phys. Rev. Lett. **45**, 814 (1980).
- ²K. O. Keshishev, A. Ya. Parshin, and A. V. Babkin, Zh. Eksp. Teor. Fiz. **80**, 716 (1981) [Sov. Phys. JETP **53**, 362 (1981)].
- ³P. E. Wolf, S. Balibar, and F. Gallet, Phys. Rev. Lett. **51**, 1366 (1983).
- ⁴P. E. Wolf, F. Gallet, S. Ballibar, and E. Rolley, J. Phys. (Paris) **46**, 1987 (1985).
- ⁵O. A. Andreeva and K. O. Keshishev, Pis'ma Zh. Eksp. Teor. Fiz. **46**, 160 (1987) [JETP Lett. **46**, 200 (1987)].
- ⁶O. A. Andreeva, K. O. Keshishev, and S. Yu. Osip'yan, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 661 (1989) [JETP Lett. **49**, 759 (1989)].
- ⁷A. V. Babkin, D. B. Kopeliovich, and A. Ya. Parshin, Zh. Eksp. Teor. Fiz. **89**, 2288 (1985) [Sov. Phys. JETP **62**, 1322 (1985)].
- ⁸S. Balibar and B. Castaing, Surf. Sci. Rep. **5**, 87 (1985).
- ⁹S. Balibar, F. Gallet, and E. Rolley, Phys. Scripta **T29**, 265 (1989).
- ¹⁰P. Nozieres and F. Gallet, J. Phys. (Paris) **48**, 353 (1987).
- ¹¹E. R. Grilly, J. Low Temp. Phys. **11**, 33 (1973).

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