Experimental Observation of a Third Roughening Transition on hcp ^{4}He Crystals

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A new roughening transition has been observed on hcp ^{4}He crystals at $T = 0.365 \pm 0.03$ K, where a third kind of facet appears at $60 \pm 3°$ from the basal planes. The authors relate this experimental observation to the arguments presented recently against the existence of quantum roughening. Some preliminary results concerning the growth of facets are also briefly mentioned.

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It is known from the work of various groups[1-5] that the equilibrium shape of the interface between hcp ^{4}He crystals and superfluid ^{4}He is not always rounded or atomically rough as was thought before, but can be partly atomically smooth, i.e., can contain facets in some crystalline directions: Two different kinds of facets appear below $T_{R1} \sim 1.15$ K (*c* or basal planes) and $T_{R2} \sim 0.85$ K (*a* planes), respectively, meaning that two different roughening transitions occur. However, various authors[6-8] assumed that these were the only two possible transitions, all the others being washed out by the quantum fluctuations of the atoms [Andreev and Parshin (A.P.)].[6] He crystals should then keep large portions of their surface rough even at $T = 0$. In a recent Letter, Fisher and Weeks (F.W.)[9] argue on the contrary that the quantum roughness does not exist and that He crystals should become completely facetted as T goes to zero. We present here evidence for the existence of a third roughening transition occurring at $T_R = 0.365 \pm 0.03$ K and concerning planes at $60 \pm 3°$ from the (0001) basal plane. These new facets correspond either to $(10\bar{1}1)$ or to $(11\bar{2}2)$ planes depending on whether *a* facets are $(10\bar{1}0)$ or $(11\bar{2}0)$; their disappearance above T_{R3} is clearly seen on growth shapes and is associated with a change measured in the size of equilibrium shapes at the same temperature. After describing our experiment, we discuss its interpretation by mentioning some preliminary results on the behavior of facets in contact with walls. Finally, we compare the hierarchy of roughening transitions found with predictions by both A.P.[6] and F.W.[9]

In order to observe He crystal surfaces, we use a dilution refrigerator with optical windows and a method similar to the one used in the "bubble" experiment by Balibar *et al.*[1] A transparent box closed at the bottom by a thin plate, in which a small hole (radius $r = 0.8$ mm) was drilled, is used to form a nearly free-standing crystal whose equilibrium shape depends on the pressure difference across the interface and the interfacial tension. The effect of gravity is small since the crystal size is less than the capillary length and the absence of contact with walls makes it easier to look at many different crystalline orientations on the interface. The hole has sharp enough edges to make the angle of contact of the bent meniscus with the bottom plate arbitrary in first approximation. It is also thin enough (0.1 mm) to avoid the complete blocking of its area by a facet as was observed in the 1978 experiment[1] and interpreted later[2] by suggesting the existence of a roughening transition around 1 K. The pressure difference across this single-crystalline bubble is obtained by measuring the height h_1 of the interface outside the box. It is monitored by adding or removing helium through the capillary to the pressure cell at constant (regulated) temperature. The height h_g of the bubble is a function of h_1, and, at constant h_1 and for some orientations, it was observed to change at the various roughening transitions, a property which provides a way to measure the various T_R at thermodynamic equilibrium. This is illustrated by Fig. 1 in the case of T_{R3}. Finally, if h_1 exceeds a critical value, the bubble becomes unstable and grows with a nearly completely facetted shape inside the box (Fig. 2). The whole crystal can be melted partially and regrown from below the hole in order to form a new bubble with exactly the same orientation.

The two photographs in Fig. 2 show two typical growth shapes obtained from the same crystal at 0.35 and 0.4 K, respectively, and under similar growth conditions. The one at 0.4 K contains a hexagonal facet which corresponds to a *c* or (0001) plane; it contains six *a* facets as well, which correspond either to $(10\bar{1}0)$ or to $(11\bar{2}0)$ planes, a point which has not been clarified yet. The appearance of these two kinds of facets on both growth and equilibrium shapes has been observed at $T_{R1} = 1.2 \pm 0.1$ K and $T_{R2} = 0.9 \pm 0.1$ K, respectively, in agreement with other experiments.[2-5]

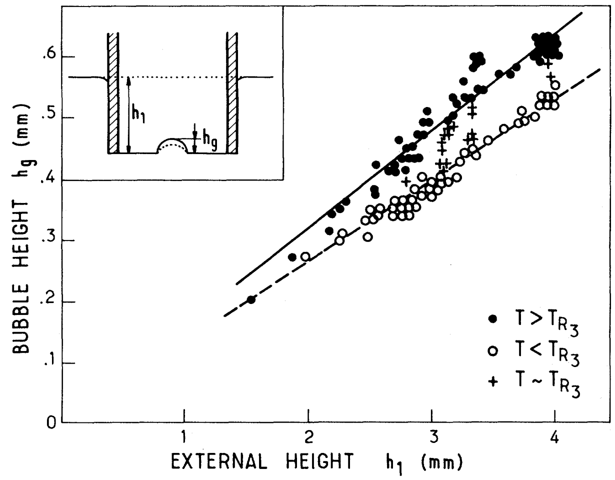

FIG. 1. The variation of the height h_g of the crystalline bubble with the height h_1 of the level outside the transparent box. Close to $T_{R3}=0.365$ K (crosses), the bubble moves from a metastable position (dotted line) to a stable one (solid line).

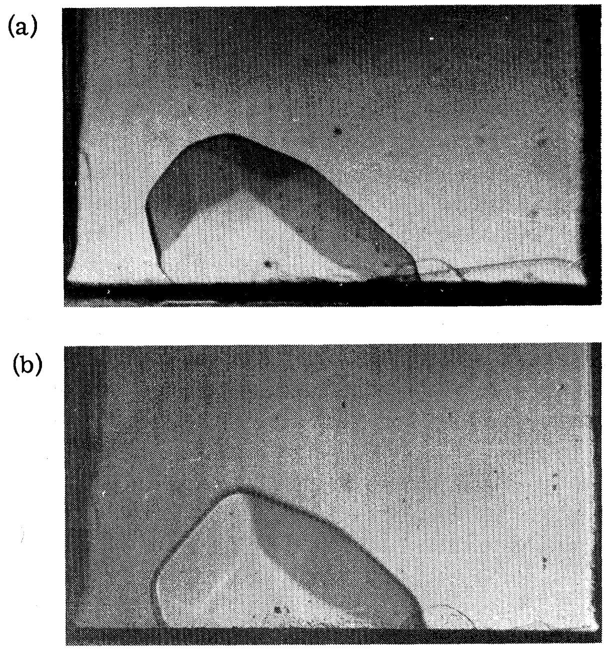

FIG. 2. Two typical growth shapes obtained in a 4-mm-wide transparent box and from the same seed: (a) at $T=0.35$ K and (b) at $T=0.4$ K. The growth velocity of facets without contact with walls is ~0.5 mm/min. Below T_{R3}, trapezoidal facets appear at 60 ± 3° from the (0001) hexagonal ones.

Note that the error bars come from the fact that these temperatures vary slightly from one crystal to another, a property which has been already noticed[4] and could be due to a varying number of dislocations or ^{3}He impurities on the interface. Our crystals were never grown at velocities larger than 1 mm/min and we use specially purified ^{4}He with a ^{3}He concentration of 1.7×10^{-9}. The precise analysis of these two first transitions will be published later and we would like to concentrate now on the third one.

The photograph at 0.35 K shows new facets, at 60 ± 3° from the basal plane, which we propose to call "*s*." These *s* facets can be either of $(10\bar{1}1)$ type or $(11\bar{2}2)$ type depending on whether the *a* ones are $(10\bar{1}0)$ or $(11\bar{2}0)$ (see Table I). All the crystals grown below 0.35 K had a shape similar to the one presented here. No other facets were ever observed on growth shapes down to 70 mK. In order to make sure that we observed a third roughening transition, and since it is always difficult to distinguish on a growth shape between zero and small curvature in a given direction, we wanted to see these facets on equilibrium shapes. We did not succeed in this experiment probably because their equilibrium size is very small. Let us remark here that even the observation of the larger *c* or *a* facets appeared difficult when the light beam was not at grazing incidence with respect to them. However, as already mentioned above, we measured a change in the bubble height h_g (Fig. 1) at the same temperature T_{R3} at which

TABLE I. A comparison between the hierarchy of roughening transitions observed and various quantities, namely the mean spacing d between parallel planes, d^2, and the maximum height d^* of the microscopic steps. a is the distance between nearest neighbors and Ω the angle between the plane and (0001).

	T_R (K)	Plane	Ω	$d\sqrt{2}/2$	d^*/a	d/d_1	$(d/d_1)^2$	d^*/d_1^*	T_R/T_{R1}
T_{R1}	1.2 ± 0.1	(0001)	0	1.155	0.816	1	1	1	1
T_{R2}	0.9 ± 0.1	$(10\bar{1}0)$	90°	0.612	0.577	0.53	0.28	0.707	0.75
		$(11\bar{2}0)$	90°	0.707	0.5	0.612	0.374	0.612	
T_{R3}	0.365 ± 0.03	$(10\bar{1}1)$	58.5°	0.541	0.637	0.468	0.219	0.78	0.3
		$(11\bar{2}2)$	62.1°	0.603	0.426	0.522	0.272	0.522	

s facets appear on growth shapes: Depending on whether the temperature is higher or lower than $T_{R3} = 0.365 \pm 0.3$ K, h_g takes a high or a low value.

The low values could only be obtained by growing the crystalline bubble from below the hole and the slow change up to the high values at a temperature T_{R3}, which varies slightly again from one crystal to another one, appeared irreversible: No change back to the low values was observed by cooling the cell. These low values correspond probably to metastable shapes with at least one s facet anchored at some place in the lower part of the hole edge. The great tendency of facets to anchor on walls has been indeed observed many times. As an example, we analyzed the growth of facets in another experiment[10]: In contrast to rough surfaces, facets grow with small velocities, a typical value being 6×10^{-4} cm s^{-1} under a chemical potential difference $\Delta\mu$ of 10 cm^2 s^{-2} across an a facet at 200 mK. Such a mobility is five orders of magnitude smaller than that of a rough surface at the same temperature.[4] However, it is not negligible, and it was observed only if the facet was completely surrounded by rough parts of the interface, i.e., without contact with walls. Facets touching walls could stay in metastable positions for hours under such small $\Delta\mu$, and move by successive jumps under an increasing $\Delta\mu$. The orientation of the crystal shown in Fig. 2 and used for all the points in Fig. 1 is such that many s facets can be anchored in the hole. When T increases up to T_{R3}, the s facets shrink, and since rough surfaces do not anchor on walls,[10] the bubble moves at $T \sim T_{R3}$ toward an equilibrium position with a higher h_g. Note that all the points at $T \neq T_{R3}$ have been measured after waiting at least 5 min more than the usual relaxation time of the bubble to its equilibrium position (2 min typically). The various points at $T \sim T_{R3}$ are not equilibrium values but are measured, on the contrary, during an observed slow motion of the interface in a few minutes. Finally, when the crystal is cooled down through T_{R3}, h_g keeps a high value because the equilibrium size of s facets is probably very small and no anchoring reappears.

Let us now examine A.P.'s and F.W.'s arguments. Note first that if we did not observe any other facet down to 70 mK, it could be due to their negligible effect on growth and equilibrium shapes but does not mean that other transitions do not exist. The observation of a "critical slowing down" by Puech *et al.*[11] around 200 mK, although referring to our work, could be related to one of these other transitions. (The difference between their transition temperature and ours is too big to be possibly interpreted by a difference in dislocation density.) According to A.P.,[6] the quantum motion can delocalize the kinks on the steps at the interface, and this leads for all crystalline planes except a few high-symmetry ones to a negative energy for isolated steps and a finite number of them at $T=0$. These zero-point steps are supposed to create a "quantum roughness" of the interface, a property which was proposed as an explanation for the fact that no facets were observed until 1979 and only two in 1980. Another presentation of the same arguments[12] consists in saying that all the roughening temperatures T_R are substantially displaced towards $T=0$, and eventually into the unphysical region $T<0$, by the effect of the zero-point motion. On the contrary, Fisher and Weeks think that A.P.'s arguments are wrong and give a lower bound for T_R:

$$T_R > (2d^2/\pi)[\tilde{\alpha}_x \tilde{\alpha}_y]^{1/2},$$

where d is the step height and $[\tilde{\alpha}_x \tilde{\alpha}_y]^{1/2}$ a mean value of the surface tension $\tilde{\alpha}$.

The main interest of our measurement is to give a hierarchy of transitions (with three values now: T_{R1}, T_{R2}, T_{R3}) which can be compared with these contradictory predictions. The exact comparison with F.W. is made difficult by the fact that the surface tension is known[1,4,13] to vary from 0.1 to 0.2 erg cm^{-2} but is not yet accurately known at T_R in the relevant directions. We then present in Table I the hierarchy of quantities d^2 only. We also compare the experimental values of T_R to the quantity d. Indeed, a simple estimate of T_R is obtained by supposing it proportional to the step energy $E_s(T=0)$. An order of magnitude for this energy is given by αn or αd, where n is the surface density of planes separated by a mean distance d and α is the surface energy. Since α is nearly isotropic,[10] this crude estimate gives T_R nearly proportional to d. A further comparison is made with a quantity d^* which is the maximum height of the microscopic steps (d^* differs from d since parallel planes are not always equidistant). This quantity d^* appears proportional to the density obtained by Drechsler[14] who allows for some surface relaxation. Estimates by Avron[3] give the same order of magnitude.

The agreement between the experimental hierarchy and the various estimates made without any quantum effect is qualitative, although out of the five planes shown to have similar densities, only three have been observed in this experiment. We have no explanation for this latter fact. Whatever it could be, there is no obvious displacement of T_{R2} or T_{R3} down to $T = 0$, and one could briefly conclude that F.W. are right. However, their arguments apply only to the ideal case of an infinite surface, for which roughness is associated with the divergence of the thermal fluctuations of the interface. In this case, the quantum fluctuations, having a finite amplitude, cannot prevent the localization of the interface by the effect of the lattice. For a real system with a finite size, the roughness has a different sense, since the thermal fluctuations keep a finite amplitude even in the rough phase. Such a roughness might be produced by quantum effects.

Finally, quantum motion might explain the very high mobilities measured for rough surfaces.[4] Since three transition temperatures are now known with some accuracy, we think that a precise calculation of the various T_R is needed. It should tell us the exact role of zero-point motion in the problem of roughening at the surface of helium crystals.

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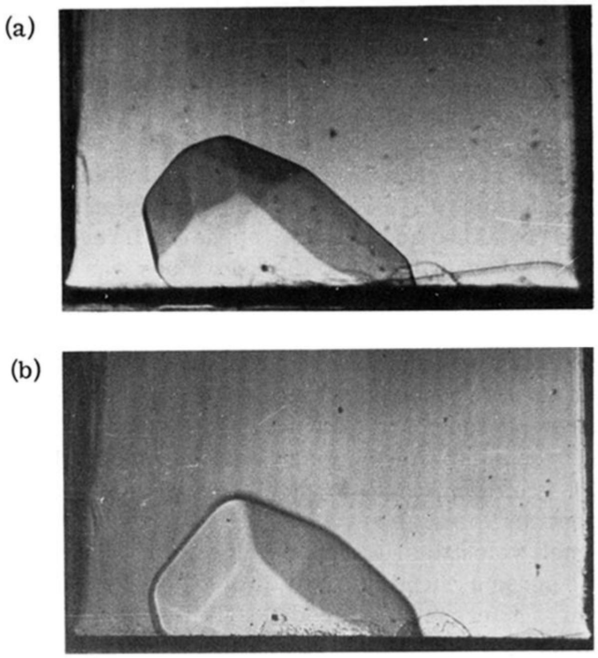

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