Crystal growth and crystal curvature near roughening transitions in hcp 4He

P.E. Wolf, F. Gallet, S. Balibar, E. Rolley, P. Nozières

To cite this version:

HAL Id: jpa-00210148
https://hal.archives-ouvertes.fr/jpa-00210148
Submitted on 1 Jan 1985

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.
Crystal growth and crystal curvature near roughening transitions in hcp $^4$He

P. E. Wolf (*), F. Gallet, S. Balibar, E. Rolley
Groupe de Physique des Solides de l'Ecole Normale Supérieure, 24 rue Lhomond, 75231 Paris Cedex 05, France
and P. Nozières
Institut Laue Langevin, B.P. 156, 38042 Grenoble Cedex,
and Collège de France, 11 place Marcellin Berthelot, 75231 Paris Cedex 05, France

(Reçu le 3 juin 1985, accepté le 4 juillet 1985)

Résumé. — Nous présentons une étude expérimentale quantitative de l'effet de la transition rugueuse sur la forme d'équilibre et la cinétique de croissance des cristaux hexagonaux d'$^4$He. D'une part nous montrons que la courbure du cristal ne s'annule pas continûment à la transition : elle présente un saut analogue à celui rencontré dans les transitions de Kosterlitz-Thouless, et la valeur correspondante de la tension de surface est en accord avec de récentes prédictions universelles. D'autre part, la cinétique de croissance des facettes devient plus rapide près de leur transition rugueuse. Dans le cas de la facette (0001), le processus dominant est la nucléation bidimensionnelle. Ceci nous permet de mesurer la variation de l'énergie de marche près de la transition. Enfin, la mobilité des facettes augmente également lorsque la température tend vers zéro, ce qui montre que la mobilité des marches cristallines est limitée par leur interaction avec les excitations thermiques.

Abstract. — We present a quantitative experimental study of the implications of the roughening transition both on the equilibrium shape and the growth kinetics of hcp $^4$He crystals. On the one hand, we show that, in close analogy with the Kosterlitz-Thouless transition, the crystal curvature does not vanish continuously at the transition. The corresponding value of the surface stiffness is found in good agreement with recent universal predictions. On the other hand, the growth rate of facets increases near their roughening temperature. For the (0001) facet, the dominant process is the two dimensional nucleation. It allows us to measure the variation of the step energy near the transition. Lastly, the facet mobility also increases when the temperature tends to zero. This shows that the mobility of crystalline steps is limited by their interaction with thermal excitations.

In 1949-1951, Burton, Cabrera and Frank predicted that, for low Miller index orientations, the surface of a crystal could change, on an atomic scale, from a low temperature smooth state to a high temperature rough one at a critical temperature $T_R$ [1]. This roughening transition has macroscopic consequences. First, since an orientation of the surface is present as a facet in the equilibrium shape of the crystal only when it is smooth, each type of facet disappears above its own roughening temperature. Second, the growth kinetics are also modified when going from the smooth to the rough state. From the theoretical point of view, the very existence of the roughening transition has been largely debated until 1976. At that time, by using a duality transformation, Chui and Weeks [2] first related this transition for a particular interface model to the Kosterlitz-Thouless transition in the two-dimensional Coulomb gas [3]. Their argument has later been extended to a wider class of interface models by Knops [4] and Jose, Kadanoff, Kirkpatrick, and Nelson [5]. Simultaneously, Van Beijeren [6] directly studied the roughening transition of the so-called BCSOS model.

However, these theoretical advances in the understanding of the transition lacked experimental support. As a matter of fact, very few crystals were known to exhibit roughening. In the case of NH$_4$Cl [7], C$_2$Cl$_6$ [7] and CBr$_4$ [8], the transition was only detected through a qualitative change in the morphology of growing crystals and could not be quantitatively studied. Metallic crystals in contact with their vapour have also been studied [9]. Despite difficulties, one can obtain the equilibrium shape of small ($\sim 10 \mu m$)
and pure specimens. For gold and lead [10], this shape presents both facets and rounded parts, but the crystal melts before any roughening transition takes place (1).

This situation changed around 1980 when the basal plane (0001) of hcp $^4$He crystals was shown to undergo a roughening transition around 1.2 K [11-13]. $^4$He crystals present many advantages for the general study of crystal growth and morphology. Since the $^4$He liquid phase is superfluid below 1.76 K and the latent heat is very small, there is no problem of heat or mass transport, in contrast with the other crystals grown from their melt. Growth kinetics are thus controlled by the interface and are fast at least for rough orientations [11]. Moreover, the $^4$He purity is orders of magnitude larger than for any other substance. This is a very important property, because the impurity adsorption on the interface drastically affects its behaviour. For these reasons, it is possible to grow large $^4$He single crystals rapidly (typically at a rate of 1 mm/min). As soon as the growth is stopped, the crystal rough surfaces relax to local equilibrium in a short time. Thus, equilibrium shapes as well as growth dynamics can be quantitatively analysed in a wide temperature range (0 → 1.76 K). This makes $^4$He an ideal system to study the roughening transition, especially since two other transitions have also been discovered in the hcp phase. The (1100) [11, 12] (rather than (1120) : see section 5) and (1101) [14] surfaces become smooth at respectively $T_R$ $\sim$ 0.9 K and $T_R$ $\sim$ 0.35 K. A fourth transition may have been observed at 0.21 K [15].

This progress has motivated in turn new theoretical works which concentrate on the equilibrium shape of crystals, peculiarly near a roughening transition. Jayaprapaksh, Saam and Teitel [17] (JST) recently pointed out that, at the transition, the crystal curvature presents a jump, which is equivalent to the superfluid density jump in the Kosterlitz-Thouless transition of $^4$He films. This result can also be arrived at from a related prediction made shortly before by Fisher and Weeks (FW) [18]. The surface stiffness $\tilde{a}(T)$ of a rough surface is given, right at the transition, by

$$\tilde{a}(T) = \frac{\pi}{2} \frac{k_B T_R}{a^2}$$

where $a$ is the spacing between crystalline planes parallel to the interface. This is an important result of the present theory of the roughening transition. As the crystal curvature is inversely proportional to the surface stiffness, it jumps at the transition from zero to a finite value. This is precisely the JST prediction. It strongly contradicts Andreev's suggestion [19] that the curvature continuously vanishes at $T_R$. In order to decide between these predictions, it appeared important to measure the surface stiffness in the rough phase, as well as the roughening temperature $T_R$.

Owing to the rapid relaxation of rough surfaces towards equilibrium, the surface stiffness can be measured by a simple capillarity experiment. As for the roughening temperature, it could be in principle determined by detecting the appearance of a facet on the equilibrium shape. However, we shall see that such a method cannot be used. On the contrary, we found that the study of growth kinetics provides a reliable determination of $T_R$, and allows us to gain a further insight in the nature of the roughening transition.

We have thus performed an experimental study of both equilibrium and growth properties of the hcp $^4$He interface, for the (0001) (« c ») and (1100) (« a ») orientations. Preliminary reports of our results have appeared elsewhere [20]. This paper is organized as follows : section 1 recalls the present understanding of the roughening transition and mainly deals with the so called « curvature jump ». Section 2 describes our experimental set-up. The growth kinetics measurements are reported in section 3. Our results for the surface stiffness in the (0001) orientation are presented and analysed in section 4, while section 5 contains all our results for the (1100) orientation. Finally, section 6 details our conclusions.

1. The roughening transition : theoretical background.

1.1 The nature of the transition. — An interface between a crystal and its fluid phase (liquid or vapour) can be, on an atomic scale, either smooth or rough according to its crystallographic orientation and temperature. A smooth interface is localized [21]. The free energy $E_a$ of a macroscopic step on this interface is then strictly positive [18, 21]. Consequently, according to the Wulff construction [22], there is a facet of corresponding orientation in the crystal equilibrium shape, whose size is proportional to $E_a$. Oppositely, in the rough state, the step energy is zero and the interface wanders arbitrarily far from any reference plane [21]. In this case, a finite size crystal is rounded in the considered orientation. The growth of the interface is also critically affected by its microscopic state. Steps and terraces exist on a rough surface at equilibrium. It can thus grow continuously in an out-of-equilibrium situation. The growth rate is then high and proportional to the chemical potential difference $\Delta \mu$ across the interface [21]. In contrast, a flat facet must overcome a macroscopic potential barrier in order to move forward by one lattice spacing. One possible mechanism is the thermally activated two dimensional (2D) nucleation (a process analogous to droplet condensation in a supersaturated 3D vapour). Near equilibrium, this process will be efficient only very close to $T_R$, when the free energy

(1) On the contrary, the disappearance of facets with increasing temperature has been observed on Ag$_2$S crystals [16].
of the critical nucleus is small. More often, growth takes place through the motion of steps issued from screw dislocations emerging on the surface: this is the well-known spiral growth [1]. In both cases, growth is usually slow and non-linear.

The possibility of a transition, for a given orientation, from smooth to rough when the temperature increases was first considered by Burton, Cabrera and Frank [1] (BCF) in the case of the (001) face of a cubic crystal. Restricting the interface excitations within one layer, they showed that the interface is microscopically flat only below a critical temperature $T_c$ (about half the nearest-neighbour interaction, in units of the Boltzmann’s constant), which they identified as the roughening temperature $T_R$. Although their argument gave a qualitative understanding of the phenomenon and facilitated discussion of the influence of roughening on both growth kinetics and crystal morphology [1, 23], it did not however rigorously prove the existence of a transition since the restriction of excitations is obviously inappropriate near $T_c$. As was shown by Van Beijeren [25], $T_c$ is then only a lower bound for $T_R$. In fact, as the roughening transition is related to the divergence of height fluctuations [21], its theory must allow the interface to extend over an infinite number of layers.

The first attempts to extend the BCF argument to multilayered interfaces were unsuccessful and casted some doubt on the very existence of the transition [1, 26]. The breakthrough occurred in 1976 when Chui and Weeks [2] used a duality transformation to relate the so-called discrete Gaussian (DG) model of the interface to a 2D Coulomb gas, which is known to present a Kosterlitz-Thouless transition [3].

Their analysis showed the crucial importance of the height-height correlation function $G(r) = \langle z(r)z(0) \rangle$, where $z(r)$ is the interface height at point $r$. In the rough phase ($T > T_R$) $G$ diverges as $\log(r/a)$, as $r$ tends to infinity ($a$ is the distance between crystal planes parallel to the interface). In the smooth phase ($T < T_R$), $G$ saturates within a distance $\xi$ to a value of order $a^2 \log\xi/a$. For a sharp interface, $\xi(T)$ is a correlation length which diverges as $\exp(c/\sqrt{T/T_c})$ and stays infinite above $T_R$ (here $\xi = \frac{T - T_R}{T_R}$). Accordingly, the step free energy $E_{\phi}$, which, as shown by Swendsen [27], is of order $k_B T_R \xi/a$, vanishes at $T_R$ as $\exp(-c/\sqrt{T/T_c})$. Thus, although one recovers the qualitative BCF picture, the universality class of the transition for the DG model is not that of the 2D Ising model, but rather that of Kosterlitz-Thouless transitions.

Shortly afterwards, José, Kadanoff, Kirkpatrick and Nelson [3], as well as Knops [4] showed that any « solid on solid » (SOS) model can be related by duality to an $XY$ model of planar spins. Moreover, at the same time, Van Beijeren [6] was able to map the SOS model for a body-centred cubic crystal (BCSOS model) into the exactly solvable six-vertex model [28].

He recovered all the results obtained by duality. These facts show that the critical behaviour at the roughening transition does not depend on the chosen microscopic model. Lastly, it must be stressed that the existence of a transition does not require the atoms to be localized on a perfect lattice, as is the case in the SOS models. A model which simply favours regularly spaced heights of the interface presents a transition with the same critical behaviour [29], a property that Chui and Weeks used to study the crystal growth near the roughening transition [30]. We shall now develop a similar model, in order to make the above considerations more precise and, perhaps, more transparent.

1.2 A SIMPLE MODEL. — We here use an approach similar to that introduced by Ohta and Jasnow [31] in their treatment of the $XY$ model. We consider an interface macroscopically normal to the direction $\mathbf{z}$ and we define an interface height $z(x,y)$ by means of an appropriate convention (for instance, the zero mass Gibbs dividing surface). We then write the energy functional for surface fluctuations as

$$ H = \int \left[ \alpha_0(1 + (Vz)^2)^{1/2} + V(z) \right] \, dx \, dy \quad (1) $$

$\alpha_0$ is the microscopic surface energy, while $V(z)$ is a potential of order $a$ which tends to pin the interface on crystal planes normal to $\mathbf{z}$. For a sharp interface, $V$ is an atomic energy (per unit cell); if, however, the liquid-solid interface extends on several atomic layers (in the absence of any fluctuations), $V$ may be much smaller. We expect this form of $H$ to hold for any low Miller index interface. Since only the first harmonic of $V(z)$ is relevant near the roughening transition [2, 29] we set

$$ V(z) = V_0 \left( 1 - \cos \frac{2\pi}{a} z \right). $$

For small fluctuations, we expand $1 + (Vz)^2 = 1 + \frac{1}{2}(Vz)^2$. We are thus left with Gaussian fluctuations coupled by the periodic potential.

Following Wilson [32], we separate fluctuations in two classes: short wavelengths ($< \ell$) and long wavelengths ($> \ell$). Averaging over short wavelength thermal fluctuations, we build an effective energy $H(l)$ for long wavelengths — hence a renormalization procedure. As long as $V$ is small we may expand $H(l)$ in powers of $V$: within second order, the energy returns the same form as (1), with renormalized coefficients $\tilde{\alpha}(l)$, $V(l)$.

$$ H(l) = \int \left[ \frac{\tilde{\alpha}(l)}{2} (Vz)^2 + V(l) \left( 1 - \cos \frac{2\pi z}{a} \right) \right] \, dx \, dy $$

(we ignore the second harmonic contribution to $V$ : for $T > T_R$, it is irrelevant as $l \to \infty$, while for $T < T_R$, it remains small as long as $l < \xi$). Note that
in \( H(l) \), all wavevectors are cut off at \( A = 1/l \). Following the algebra of Ohta and Jasnow, we obtain the recursion equations

\[
\frac{dU}{d\eta} = U \left(2 - \frac{\pi k_B T}{\alpha a^2} \right) \tag{2}
\]

\[
\frac{d\tilde{\alpha}}{d\eta} = \frac{2 A^2 U^2}{\tilde{\alpha} a^4} \tag{3}
\]

in which \( \eta = \log \frac{1}{\alpha_0} \) (\( \alpha_0 \) is the microscopic cut off, of the order of the interface width). \( U = V l^2 \) is the effective potential energy on the new unit cell (of area \( l^2 \)), to be compared to \( k_B T \). \( A^2 \) is a non universal constant, which, for a square cut off, takes the form

\[ A^2 = \pi^4 \int_0^\infty \frac{\rho^3 d\rho}{l^3} J_0(\rho/l) \simeq \pi^4/4. \]

(This value of \( A^2 \) is also obtained for many other possible cut-off [31]). Note that the denominator of (3) involves \( \tilde{\alpha}(l) \), instead of the bare \( \alpha_0 \), as in Ohta and Jasnow’s case. The scaling equations (2) and (3) contain all the physics of the problem. The microscopic behaviour corresponds to \( l \to \infty \) : the limit \( \tilde{\alpha}(\infty) \) of \( \tilde{\alpha}(l) \) is nothing but the macroscopic interface stiffness \( \tilde{\alpha} \) for the interface normal to \( l \) (where \( \tilde{\alpha} \) is defined from the surface energy \( \alpha(\phi) \) by the usual relation \( \tilde{\alpha} = \alpha + \frac{\partial^2 \alpha}{\partial \phi^2} \), where \( \phi \) is the crystallographic angle with respect to the \( z \) direction).

Let us define dimensionless quantities

\[
X = 2 \left( \frac{2 \tilde{\alpha} a^2}{k_B T} - 1 \right), \quad Y = \frac{4 A U}{k_B T} \]

in terms of which (2) and (3) become

\[
\frac{dX}{d\eta} = \frac{Y^2}{1 + X/2} \tag{4}
\]

\[
\frac{dY}{d\eta} = \frac{XY}{1 + X/2}. \tag{5}
\]

The scaling trajectories are hyperbolae \( X^2 - Y^2 = C_o \).

At any temperature, scaling starts when \( \eta = 0 \) at some initial point \( X_0, Y_0 \), whose locus, as \( T \) is varied, is a straight line (Fig. 1).

\[
Y_0 = t_e (1 + X_0/2), \quad t_e = \frac{2 AU_0}{\alpha_0 a^2} \]

\( t_e \) is the only parameter of the problem; it measures the strength of the original potential \( V_0 \) and must be much smaller than 1 if the perturbation calculation is to be valid.

The roughening temperature corresponds to \( C_0 = 0 \) (see below, (i) and (ii)) and hence \( X_0 = Y_0 = 0 \), or

\[
k_B T_R = \frac{2 \alpha_0 a^2}{\pi} \left(1 + \frac{t_e}{2}\right).
\]

At the transition, the macroscopic surface stiffness is given by the condition \( X = 0 \):

\[
\tilde{\alpha}(T_R) = \frac{\alpha_0}{2} a^2. \tag{6}
\]

The relation between \( \tilde{\alpha}(T_R) \) and \( T_R \) is thus universal [18, 29, 30] : it does not explicitly depend on the microscopic interactions between the atoms or, more generally, on the chosen microscopic model (3). In the vicinity of \( T_R \), we introduce the reduced temperature \( t = (T - T_R)/T_R \). To the extent that \( t, t_e \ll 1 \), it is easily found that \( C_0 \simeq 4(t + t_e) \). We then expect the following behaviour :

(i) \( T > T_R \) : Scaling drives \( Y \) to 0 : the pinning energy \( U = V l^2 \), becomes irrelevant as \( l \to \infty \). The interface thus fluctuates freely : it is rough. The macroscopic surface stiffness achieved when \( l \to \infty \) is given by

\[
\tilde{\alpha}(T) - \frac{\alpha_0}{2} t + \frac{t_e}{2} - \sqrt{t(t + t_e)} \quad (t, t_e \ll 1). \tag{7}
\]

In the critical region \( t \ll t_e \), it approaches \( \tilde{\alpha}(T_R) \)

\[ (3) \quad \text{This relation implies that the correlation function} \quad G(r) \quad \text{behaves as} \quad \frac{2}{\pi^2} \log \frac{r}{\alpha} \quad \text{right at the transition. This property has been used in numerical simulations to determine the value of} \quad T_R \quad \text{see reference [36].} \]
as $\sqrt{t}$. For $t \gg t_c$, $\bar{a}(T)$ is equal to its bare value $a_0$ (*): scaling does not affect the surface stiffness (the hyperbola reduces to its vertical tangent). Note that there is no characteristic length in this case: the correlation length is infinite.

(ii) $-T < T_R$, $|t| \ll t_c \ll 1$. $Y$ first decreases and then increases until the perturbation expansion breaks down. The periodic potential becomes statistically significant when $U/T \sim 1 / \log |t|$ ($Y \sim A$). The corresponding length scale $\xi$ marks the correlation length, beyond which the interface is pinned. The interface is thus localized on a scale larger than $\xi$ while it looks rough on a smaller scale: vertical height fluctuations are of the order of $a \left( \frac{\log \xi}{a} \right)^{1/2}$. Physically, $\xi$ is the typical size of thermal fluctuations (i.e. thermally excited terraces and holes). $\xi$ also corresponds to the thickness of a macroscopic step. This thickness can be thought of as the typical transverse extension of the fluctuating microscopic steps. From $\xi$, we then infer the step free energy $E_s \sim \bar{a}(\xi) \xi^2 / \xi \sim kT_0 / \xi$. The integration of the scaling equations yields the Kosterlitz result [3]:

$$\xi / a_0 \sim \exp \left( \frac{\pi}{2 \sqrt{|t| / t_c}} \right).$$

Near $T_R$, $\xi$ diverges very rapidly. Thus, as one approaches $T_R$, a macroscopic step broadens and ultimately vanishes, while its free energy goes to zero.

(iii) $T < T_R$, $t_c \ll |t| \ll 1$. In this regime, which exists only for very small $t_c$, the correlation length grows slower than in the case (ii):

$$\xi / a_0 \sim \exp \left( \frac{\log (4 |t| / t_c)}{2 |t|} \right).$$

The above analysis dealt with an interface which is, on the average, parallel to the crystal planes. However, a real experiment only measures some local average of $\bar{a}(\phi)$ near $\phi = 0$. Therefore, it is important to determine the thermal evolution of the surface stiffness of a surface tilted by an angle $\phi$. We thus now consider the effect of a non-zero average slope on the renormalization process. A similar situation was considered by Horovitz et al. [33] in the related problem of incommensurate transitions. Translated to our context, their results show that the renormalization is essentially stopped when the length scale $l$ is comparable to the distance $a/\phi$ along the interface, between the successive crystalline planes, this fluctuation averages out the periodic potential. Hence, it does not contribute to the renormalization of $\bar{a}(\phi)$. This result can also be obtained in our language. Putting the tilt by hand in the periodic potential, we write the energy of a horizontal surface making a small angle $\phi$ with the crystalline planes as

$$H = \int \left\{ \frac{a_0}{2} \nabla \phi \right\}^2 + V_0 \left( 1 - \cos \frac{2\pi}{a}(\phi - \phi_0) \right) \times d\phi d\rho.$$  

We then eliminate the short wavelength fluctuations as before. The resulting scaling equations are unchanged, except for the expression of $A^2$, which becomes

$$A^2 = \frac{\pi^3}{2} \int \frac{\rho^2}{4\pi^2} J_0 \left( \frac{\rho}{\xi} \right) \cos \frac{2\pi \rho \phi}{a} d\rho.$$  

As long as $l \ll a/\phi = d$, the cosine is 1 and $A^2$ is the same constant as in the $\phi = 0$ case: logarithmic scaling proceeds without any noticeable effect of the tilt. On the other hand, when $l > d$, the cosine averages $A^2$ to zero, so that the surface stiffness $\bar{a}$ does not change any longer, while the periodic potential becomes irrelevant, since it oscillates rapidly on the scale $l$ (\(5\)).

We can thus infer the macroscopic behaviour of the surface stiffness $\bar{a}(\phi)$ by integrating our original scaling equations up to $\eta = |\log \phi|$. The algebra is straightforward, but somewhat clumsy: we only quote a few salient results, valid for $t, t_c \ll 1$.

(i) At $T = T_R$, we get

$$\bar{a}(\phi) - \bar{a}^* = - \frac{t_c}{\log |\phi|} \left( \bar{a}^* - \bar{a}(0) / \bar{a}(0) \right),$$

where $\bar{a}^* = \bar{a}(T_R, \phi = 0)$ is the universal surface stiffness at the transition. The approach to $\bar{a}^*$ when $\phi$ tends to zero is very slow (the logarithmic behaviour was found by Horovitz et al. [33]). When $\phi$ exceeds a value of order $\exp(-1/t_c)$, $\bar{a}(\phi)$ practically returns to the bare value $a_0$: the effect of the potential is negligible. When $t_c \ll 1$ (a condition for our perturbation method to be valid), the angular domain in which the critical behaviour is visible is thus extremely narrow.

(ii) In the high temperature critical region, $0 < t \ll t_c$, the difference $|\bar{a}(\phi) - \bar{a}(0)|$ is essentially the

(*) Strictly speaking, the surface stiffness $\bar{a}$ becomes anisotropic in the transition region: $\bar{a}_x \neq \bar{a}_y$. One may write separate scaling equations for $a_x$ and $a_y$, which are non-logarithmic—this is the procedure followed by Horovitz et al. [33]. Within logarithmic accuracy, such a refinement is not needed. It is simpler to stop scaling when $l \sim d$. 

(*) This is true because, in the whole discussion, we have ignored the effect of the « free » thermal fluctuations on a. As long as one deals with the critical behaviour at $T_R$, it is justified, since this effect does not lead to any singularity. However, if taken into account, this effect would add to equation (7), a term linearly decreasing with temperature (in the classical limit).
same as at $T = T_R$ as long as $\phi > \exp \left[ -\frac{1}{4\sqrt{t_c}} \right]$.

For smaller angles, the dependence of $\tilde{\sigma}(\phi)$ on $\phi$ is given by the power law

$$\frac{\tilde{\sigma}(\phi) - \tilde{\sigma}(0)}{\tilde{\sigma}^*} \sim - \frac{1}{2\sqrt{t_c}} \phi^{4/3}$$

which is hardly observable in practice. In the opposite limit $t \gg t_c$, we again find a power law

$$\frac{\tilde{\sigma}(\phi) - \tilde{\sigma}(0)}{\tilde{\sigma}^*} \sim - \frac{t^2}{8t} \phi^{4t}.$$ 

In both limits, $\tilde{\sigma}(\phi)$ returns to its bare value $\sigma_0$ as soon as $\phi$ is large enough. Renormalization effects can only be seen at very small angles $\phi$ (unless $t_c \gg 1$, in which case our discussion is only qualitative, because the perturbation expansion is not really valid).

(iii) When $T < T_R$, the central question is whether one should stop scaling before or after the renormalization process diverges, i.e. whether $d = a/\phi$ is smaller or larger than the correlation length $\xi$. In the former case, $\phi < a/\xi$, the tilt does not affect the calculation much: the surface is built with well separated steps. In the latter case, $\phi > a/\xi$, one stops scaling before the steps have time to develop. Put another way, steps are so close $(d < \xi)$ that they overlap, thereby losing their identity. The corresponding surface stiffness $\tilde{\sigma}(\phi)$ can again be obtained by integrating the scaling equations. In the critical regime $|t| \ll t_c$, $\tilde{\sigma}(\phi)$ is nearly the same as at $T = T_R$ as long as $\phi > a/\xi$ (going from $\sigma_0$ at large angles to $\tilde{\sigma}^*$ when $\phi \leq \exp - 1/t_c$). After a flat plateau, it grows until $\phi \sim a/\xi$, where the scaling perturbation breaks down. Further away from the transition ($|t| \gg t_c$), the plateau disappears.

In the region of well developed steps $(\phi < a/\xi)$, the surface energy is, in lowest order, proportional to the step density $n : f = nE_s$ per unit area parallel to the crystal planes. Corrections arise from step interactions due partly to step fluctuations [34], partly to an elastic coupling via the deformation of the crystal [35]. In both cases, interactions go as $1/d^2$, resulting in a surface energy

$$f(n) = \sigma_0 + E_s n + \gamma n^3.$$ 

It is easily shown that $\tilde{\sigma} \sim d^2 f / \partial n^2$. Hence, as long as the step picture holds (i.e. when $\phi < a/\xi$), the surface stiffness of a vicinal surface varies linearly with angle $(\tilde{\sigma} \sim \gamma \phi)$.

The former considerations allow us to sketch in figure 2 the behaviour of $\tilde{\sigma}(\phi)$ for various temperatures. When $T \gg T_R$, $\tilde{\sigma}$ decreases at low $\phi$, in a narrow angular range if $t_c < 1$: the critical behaviour at $\phi = 0$ is rapidly washed out when $\phi$ is finite. When $T < T_R$, we only know the behaviour of $\tilde{\sigma}$ at small and large angles $\phi$, which suggests that $\tilde{\sigma}$ goes through a maximum for $\phi \sim a/\xi$. Estimating this maximum is beyond reach of the present approach.

1.3 Consequences on macroscopic quantities. — Until 1980, the above analysis (for $\phi = 0$) was supported by some numerical calculations [36-37], but an experimental test of the theoretical predictions was still lacking. As mentioned in the introduction, the discovery of three roughening transitions in hcp $^4$He crystals has then renewed the interest in the field. It has partly motivated recent works which dealt with the macroscopic consequences of the roughening transition on the crystal equilibrium shape. By using a connection, due to Andreev [19], between this shape — for a free-standing crystal — and a Legendre transform of the interfacial free energy $f$, Jayaprakash, Saam and Teitel made important predictions [17].
We can derive their results from the analysis carried out in § 1.2 and the relation between the principal curvature radii of the surface of a free crystal and the corresponding surface stiffness:

\[ R_i = \frac{2 \tilde{\alpha}_i}{\Delta P} \quad i = 1, 2 \]  \hspace{1cm} (9)

where \( \Delta P = \rho_s - \rho_l \) \((\rho_s - \rho_l)\). \( \rho_l \) and \( \rho_s \) are the liquid and solid densities; \((P_1 - P_0)\) is the excess pressure of the liquid with respect to the equilibrium pressure (at the same temperature) of a flat interface (6) and depends on the crystal size. This relation (9) can be established by using, for example, Andreev’s formalism. Now, according to equation (6), \( \tilde{\alpha}(T_R) = \frac{\pi}{2 a^2 k_B T_R} \), so that the surface stiffness remains finite at the transition, a result (7) first emphasized by Fisher and Weeks [18]. If we allow for a possible anisotropy of the surface stiffness tensor, equation (6) remains valid if \( \tilde{\alpha} \) is replaced by the geometrical mean \((\tilde{\alpha}_1 \tilde{\alpha}_2)^{1/2}\) of the two principal surface stiffnesses [18]. In terms of the crystal principal curvatures, (6) then translates into

\[ (C_1 C_2)^{1/2} = \frac{\Delta P a^2}{\pi k_B T_R} \]  \hspace{1cm} (10)

which is precisely the JST result (as their Lagrange parameter \( \lambda \) [17, 19] is \( \Delta P/2 \)). At the roughening transition, the crystal curvature thus displays a discontinuous \textit{jump}, going from the universal value \( 2 \tilde{\alpha}(T_R) \Delta P \) just above \( T_R \) to zero just below, when the flat facet appears. Since the facet radius is zero at \( T_R \), the jump only occurs at one point and the overall crystal shape varies continuously. Note that the curvature jump, as the value \( \tilde{\alpha}(T_R) \), is universal.

Below \( T_R \), assuming an isotropic step energy the equilibrium facet radius \( L \) is given by (8)

\[ L = \frac{2 E_s}{a \Delta P}, \]  \hspace{1cm} (11)

Since \( E_s \sim \bar{a}^2/\xi \), the facet size should vanish at \( T_R \) in the non analytic fashion (for a given \( \Delta P \)):

\[ L \sim \exp \left[ -\frac{\pi}{2(t | t_c |)^{1/2}} \right] \quad (| t | \ll t_c \ll 1). \]  \hspace{1cm} (12)

The same \( t_c \) enters the critical behaviour of the surface stiffness above \( T_R \) (cf. Eq. (7))

\[ \tilde{\alpha}(T) = \tilde{\alpha}^*(1 - (tt_c)^{1/2}) \quad (t \ll t_c \ll 1). \]

Hence, in principle, it is possible to check the consistency of the theory by measuring the critical behaviour on both sides of the transition.

The above behaviour, characteristic of the Kosterlitz-Thouless transition, should be contrasted with a previous theory by Andreev [19], according to which the curvature should linearly vanish at \( T_R \):

\[ \frac{1}{R} \sim (T - T_R) \]  \hspace{1cm} while the facet radius should behave as \( (T_R - T)^{1/2} \). These different predictions are in striking disagreement: experiment should distinguish between the two models.

To conclude this connection to the equilibrium crystal shape, let us mention that, below \( T_R \), the behaviour \( \tilde{\alpha}(\phi) \sim \gamma \phi \) (§ 1.2) implies that, near the facet edge \( (x = x_0) \), the crystal profile \( z(x) \) is given by \( z(x) \sim (x - x_0)^{3/2} \), as was first shown by JST [17]. However, this behaviour should only occur when the steps are well separated, i.e. for angles \( \phi \) less than \( \alpha/\xi \), that is in a vanishingly small angular domain when \( T \to T_R \).

2. Experimental set-up.

In this section, we first present the general characteristics of the refrigerators in which we obtain and observe \( ^4 \text{He} \) crystals. We then describe the experiments used to measure the surface stiffness and the growth kinetics of the (0001) and (1100) orientations.

The \( c \)-facet was studied down to 0.8 K in a pure \( ^4 \text{He} \) optical cryostat. Crystals are observed through four windows separated by vacuum and indium-sealed on the 300 K, 77 K, 4 K shields and the pressure cell (Fig. 3). For better mechanical strength-and thermal conductivity, the cell windows are made out of sapphire. Pyrex glass is used for the other windows, since it absorbs thermal 300 K infrared radiation better than fused silica. The smallest window has a diameter of 30 mm, so that the field of observation is quite large. We have four series of such windows, allowing to observe in two right-angle directions. Total heat losses due to infrared radiation through the windows are less than 100 mW. An optical dilution cryostat was used to perform experiments on the \( a \)-facet between 70 mK and 1 K. It has only two series of six windows, so that observation is only possible in one direction. The window sizes are about the same as in the \( ^4 \text{He} \) cryostat. Heat losses due to infrared radia-
tion amount to ~ 50 µW. In both cryostats, the temperature is regulated to better than 5 × 10⁻⁴ K. It is measured by four-terminal germanium resistors in contact with superfluid. These have been calibrated by using standard CMN thermometry, starting from the ⁴He saturated vapour pressure curve. The temperature is finally determined with a precision of about 1%. Crystals are grown at constant temperature in the pressure cell (diameter 60 mm, height 90 mm) by adding or removing purified ⁴He through a capillary. (The ³He concentration is 1.7 × 10⁻⁹). It will appear that we need the studied surface to be horizontal. We thus orient the crystals by using a technique first developed in Moscow [11]. Two 50 µm diameter insulated copper wires are bifilarly wound on a 5 mm diameter, 10 mm long plexiglass rod. The resulting capacitor is suspended in the cell. By applying a voltage difference of 500 V between the wires, we create an electric field of several hundreds of kV/cm near the capacitor. This gives rise to a difference of chemical potentials between the solid and the liquid, because the solid electric permittivity is higher than the liquid one. This difference has the same order of magnitude as that corresponding to the 10⁻² bars excess pressure found necessary to nucleate ⁴He crystals in the absence of any electric field [38]. We thus expect the crystal to nucleate on the capacitor, which is indeed observed. As soon as the crystal is large enough (several cubic millimeters), it falls to the bottom of the cell. When this occurs around 1 K, only the (0001) orientations are faceted. The crystal then grows as a flat object, which « glides » through the cell during its fall and sometimes orients with the large facets being horizontal. In this way, we get crystals whose c-axis is close to vertical (1 or 2 degrees away at most). On the above described capacitor, several crystals nucleate at the same time, but once they have fallen down, the large one usually grows while the others melt. The problem can anyway be avoided by using a smaller capacitor, in which case only one crystal is formed. Once a properly oriented crystal is obtained, we melt it down to a size of about one cubic millimeter in order to get the best possible seed for the growth of a larger crystal. By applying the same method below TR², crystals with an horizontal a-facet can also be obtained, although with more difficulty.

We measure the surface stiffness by using a method similar to the one described in reference [39]. A transparent box (5 mm × 5 mm wide) is suspended in the cell. Liquid ⁴He can flow in and out of the box through its open top. The bottom plate is a thin (0.05 mm thick) copper foil. A 0.83 mm diameter hole is drilled through its centre. When we grow a crystal, it first spreads on the bottom of the cell and finally fills its lower part. Since the capillary length is about one millimeter, the interface is flat, except close to the cell walls. As more helium is added, the interface level rises. When it is slightly above the bottom of the box, the interface inside the box forms a small meniscus resting on the hole edges, as in figure 4. We recall that solid ⁴He does not wet walls [38]. The hydrostatic pressure difference between the two sides of the meniscus is balanced by the surface stiffness, so that (9)

\[
\frac{\varrho_1}{R_1} + \frac{\varrho_2}{R_2} = \Delta \varrho h .
\]

Here, \(R_1\) and \(R_2\) are the principal curvature radii at a given point of the meniscus. \(h\) is the height difference between this point and the flat outer level and

\(\text{Fig. 4. — The geometry used for measuring the surface stiffness. The scale is given by the visible width of the box, which is 3 millimeters. The dark horizontal line in the upper part of the photograph is the } ^4\text{He solid-liquid interface outside the box. Inside the box, the interface forms a small meniscus which rests on the edges of a hole drilled through the bottom plate. Measurements of the outer level height and of the meniscus local curvature yield the surface stiffness.}

\(\text{(9)}\)\) Strictly speaking, this is a consequence of phase equilibrium rather than mechanical equilibrium: see note \((\ast)\).
\[ \Delta \rho = \rho_s - \rho_l \] [40]. For a vertical \( c \)-axis, and since the hole is circular, \( R_1 \) and \( R_2 \) are equal at the top of the meniscus. The surface stiffness in the (0001) direction is then given by

\[ \tilde{a} = \Delta \rho g H R/2 \] (14)

\( H \) being the value of \( h \) at the top of the meniscus and \( R \) the curvature radius of any normal section of the surface containing the \( c \)-axis. According to equation (9), this is the same result as for a free standing crystal. The case of a vertical \( a \)-axis is more complicated and will be discussed in section 5.

When the outer level rises, \( H \) increases and \( R \) decreases. Since the meniscus standing on the hole cannot have an arbitrarily small curvature, the Laplace equation can be fulfilled only below some critical level difference \( H_c \). If this difference is exceeded, the meniscus becomes unstable and begins to grow. The photograph of figure 5 shows this phenomenon for a vertical \( c \)-axis: at a temperature between \( T_{R1} \) and \( T_{R2} \), the meniscus first fills the whole box width, since the horizontal \( c \)-facet grows slower than the other orientations. Note that, as no more helium is added to the cell once the growth begins, the outer level remains fixed and at equilibrium (or very near: see below). If there is no threshold for the facet growth, the inner interface then relaxes, at a rate controlled by the facet kinetics, to its equilibrium position (which is lower than the outside interface by the capillary depression \( H_0 \)). The departure from equilibrium is best expressed by the chemical potential difference per unit mass \( \Delta \mu = \mu_l(P_l, T) - \mu_l(P_0(T)) = (\rho_l^{-1} - \rho_s^{-1})(P_l - P_0(T)) \), where \( P_l \) is the liquid pressure at the interface and \( P_0(T) \) the equilibrium pressure of a flat interface [10]. Let us now express this difference as a function of \( H \), the level difference between the inner and outer interfaces. Because the growth velocity \( V \) is small, the temperature gradient due to the released latent heat is negligible, so that the inner and outer interfaces are at the same temperature. Moreover, as the liquid flow towards the growing interface varies only slowly with time, the variation of the liquid pressure is hydrostatic. \( \Delta \mu \) is then given by

\[ \Delta \mu = \frac{\rho_l - \rho_s}{\rho_s} gH. \] (15)

Strictly speaking, this should be increased by the chemical potential difference \( \Delta \mu^{\text{ext}} \) across the outer interface. \( \Delta \mu^{\text{ext}} \) arises from the necessary melting of the outer interface (the experiment is carried out at a constant total mass) and also from the temperature (or pressure) fluctuations. However, the outer interface is at least hundreds times larger than the inner one and is largely rough (near the walls). It then absorbs the fluctuating part of \( \Delta \mu^{\text{ext}} \) in a very short time compared to the response time of the inner interface (11). There only remains the part of \( \Delta \mu^{\text{ext}} \) due to melting, which is much less than \( \Delta \mu \), again because of the difference of sizes (and hence of velocities) between the two interfaces. Hence, \( \Delta \mu^{\text{ext}} \) can be neglected and we do get expression (15). The relation \( \nu(\Delta \mu) \) is thus obtained by measuring \( H \) as a function of time. The experiment is carried out as follows. We raise the outer level to its critical position, allow the temperature to stabilize and

---

(10) This choice, which avoids any reference to the ill-defined solid pressure, is appropriate for a macroscopically flat interface. However, for a curved rough interface, the actual chemical potential difference across the interface is \( \Delta \mu = \mu_l(P_l, T) - \mu_l(P_0 + \Delta P, T) \) with \( \Delta P = \frac{\tilde{a}_1}{R_1} + \frac{\tilde{a}_2}{R_2} \). Accordingly, due to the non zero curvature of a rough interface in the box, its equilibrium position is given by \( \Delta \mu^* = \frac{\Delta \rho}{\rho_s} g(H - H_0) = 0 \), i.e. \( H = H_0 \), the capillary depression but not by \( \Delta \mu = \frac{\Delta \rho}{\rho_s} gH = 0 \).

(11) This rapid relaxation of the outer interface towards equilibrium is very important. Otherwise, a \( 10^{-4} \) K temperature fluctuation away from the equilibrium curve would be equivalent at 1 K to an additional overpressure of about \( 4 \times 10^{-5} \) atm, or to an extra height \( H = 2 \) mm, which is quite large. This role of « reservoir » played by the outer level is experimentally demonstrated by the temporal stability of the meniscus studied in section 4. On the contrary, a small crystal alone in the cell keeps oscillating back and forth because of thermal or mass fluctuations.
then add a tiny amount of $^4$He to destabilize the meniscus without changing the temperature. The further growth is measured with a cathetometer, or, when too fast, recorded on a video tape. The highest value of $H$ is generally the critical height, which is about 5 mm for the 0.83 mm diameter hole used. On the other hand, this method does not allow precise measurements of $H$ below 0.5 mm, because the curvature of the outer interface makes then focusing difficult. We have thus studied growth in a range of values of $H$ from 0.5 up to 5 millimeters. This corresponds to liquid overpressures $P_l - P_o = 10^{-5} - 10^{-4}$ atm or to $\frac{\Delta \mu}{k_B}$ (per atom) $\sim 10^{-7} - 10^{-6}$ K.


Figure 6 presents the growth velocity $V = \frac{dH}{dt}$ as a function of $H$ for several temperatures in the neighborhood of the roughening temperature previously estimated as $T_R = 1.19$ K [20, 41]. As is expected, the relation $V(\Delta \mu)$ strongly depends on temperature. Above about 1.23 K, the velocity is linearly related to $H$. This is demonstrated by the semi-logaritmic plot of figure 7, which shows that $H$ relaxes exponentially with time to $H_o$, the capillary depression in the box (a crude calculation gives $H_o$ of the order of 0.2 mm for the 5 x 5 millimeter box). According to note (10), $V$ is thus proportional to the chemical potential difference across the interface $\Delta \mu^* = \frac{\Delta \mu}{\rho_s} g(H - H_o)$.

This is not inconsistent with a rough state of the interface above 1.23 K. Moreover, above this temperature, the mobility $k = V/\Delta \mu^*$ is only slightly temperature dependent, and is about $6 \times 10^{-3}$ s/cm, which is close to the mobility of rough orientations [42] at the same temperature. On the contrary, below 1.23 K, the relation $V(H)$ is no longer linear. Furthermore, as shown in figure 8, for any given value of $H$, the velocity $V(H)$ strongly decreases with temperature. This is exactly what is expected for a smooth surface which grows layer by layer. Thus, the roughening temperature is at least 1.23 K (12).

In order to go further, we first need to know the growth mechanism near the roughening temperature. The analysis of our whole set of results shows that the relation $V(\Delta \mu)$ is reproducible from one crystal to another, at a given temperature. Moreover, it is highly non linear (this appears more clearly in figure 9, where points with velocities smaller than one micron per second are also added). These two facts suggest that the facet growth does not proceed by spiral growth (in which case $V$ should depend on $\Delta \mu$ at most quadratically [1]), but by two dimensional nucleation.

Although we are studying the growth very near equilibrium ($\Delta \mu/k_B T :\leq 10^{-5}$), 2D nucleation could indeed occur since the step energy should be very small close to $T_R$. In order to test this hypothesis, let us first recall the expected relation $V(\Delta \mu)$ in this case. If we assume an isotropic step energy, the energy of the critical 2D nucleus is $E = \frac{\pi E_s^2}{2(\Delta \mu \rho_s)}$. The nucleation rate is then proportional to $\exp(-E/k_B T)$. As shown for instance in reference [21], the growth velocity $V$ varies as the one third power of this rate, i.e. as $\exp(-H^*/H)$ with $H^*(T) = \frac{\pi E_s^2}{3 a \Delta \mu \rho_s k_B T}$. There is also a prefactor which includes the step mobility and varies as $H^{5/6}$. We can then approximately expect $V \sim H \exp(-H^*/H)$ for a 2D nucleation process.

(12) This value 1.23 K is higher than the value 1.20 K used in reference [20]. This modification is due to recent and more precise measurements of the growth kinetics at low values of $\frac{\Delta \mu}{k_B T}$. 

---

Fig. 6. — The growth velocity $V$ of the c-orientation as a function of the level difference $H$, for various temperatures near the roughening transition. Above 1.23 K, $V$ is a linear function of $H$ (see also the Fig. 7). This is not the case below 1.23 K, which indicates that the roughening temperature is at least 1.23 K.

Fig. 7. — Semi-logarithmic plot of $H(t) - H_o$ versus time $t$. (The capillary depression $H_o$ is taken as 0.1 mm.)
Fig. 8. — Ratio of the growth velocity $V$ of the c-orientation to the level difference $H$ ($H = 3$ mm), as a function of temperature. Different symbols refer to different crystals. Note the sharp decrease of $V/H$ below 1.23 K. Above 1.23 K, $V$ is proportional to $H$ and the ratio $V/H$ is proportional to the mobility of the surface. The dashed lines represent measurements of $V/H$ for other rough orientations [42].

We have thus plotted in figure 9 $\log \left( \frac{V}{H} \right)$ as a function of $1/H$ for the studied temperatures. The different symbols refer to different crystals or runs. We see that, at any temperature, $\log V/H$ is indeed a linear and reproducible function of $1/H$. This agreement, which does not use any adjustable parameter, extends on the whole range of velocities, i.e. on more than two orders of magnitude in the best cases. Note that the lowest values of $V/H$ correspond to velocities of about $10^{-2}$ microns per second and thus require long observation times. As the temperature increases, the slope $H^*$ goes continuously to zero. This is consistent with its proportionality to the square of the step energy. We thus believe that figure 9 shows that the growth of the (0001) facet does occur by 2D nucleation near $T_R$.

Above 1.23 K, the slope is zero within the experimental error, which means, as already mentioned, that, in our experimental range of $H$ values, $V$ is proportional to $H - H_0 \approx H$ only above this temperature.

We finally remark that, at high values of $H$ ($H \gg H^*$), $V/H$ tends to a limit $5-10 \times 10^{-2}$ s$^{-1}$, whatever the temperature. In some sense, this is expected. Indeed, near $T_R$, the ratio $H^*/H = \frac{\pi E_s R^*}{3 k T}$ (where $R^*$ is the critical nucleus radius) is of order $R^*/\xi$, where $\xi$ is the step thickness. Hence, for $H \gg H^*$, $R^*$ is much smaller than $\xi$ and critical nuclei are spontaneously present on the surface. Put in another way, the smooth surface grows as it were rough, and $V/H$ depends only weakly on temperature. However, it is striking that this limit is reached as a direct extrapolation of the low $H$ regime, where $V/H \sim \exp(-H^*/H)$. This point shows that the mobility of steps, which controls the prefactor of $\exp(-H^*/H)$ and the mobility of rough surfaces are at least of the same order of magnitude. Thus, for a moving rough interface, the dissipation due to rotons is mainly controlled by their collisions with the surface kinks [13].

We can now estimate the step energy from the relation $H^* = \frac{\pi E_s \xi}{3 a \Delta p k_b T}$. Its temperature variation is shown in figure 10. We first note that a square root dependence of $E_s/a$ such as predicted by Andreev [19] is inconsistent with our results. A typical value of $E_s/a$ in the studied temperature range is $2 \times 10^{-4}$ erg/cm$^2$, that is about 1 000 times smaller than the

(13) This idea was first mentioned to us by D. O. Edwards and is also present in the work of Kosevich and Kosevich [43]. It should be opposed to that of Andreev and Parshin [44] (or Andreev and Knizhnik [45]) according to which the dissipation would mainly be a « bulk » effect. Unlike the latter, this idea explains why the rough surface mobility is not affected [46, 47] when the transport of rotons becomes hydrodynamic. Indeed, even then, although the drift velocity of the roton gas is equal to that of the interface, the relative velocity of rotons and kinks cannot be zero (another explanation of the effect has also been presented by Castaing [47]).
Fig. 10. — Temperature dependence of the step energy \( E_s \), as deduced from figure 9 and other similar measurements. Error bars have about the symbols size. \( E_s/a \) is much smaller than the surface stiffness \( \tilde{\alpha} \approx 0.2 \) erg/cm\(^2\) and vanishes, within the experimental precision, near 1.23 K. Comparison with the present theory of roughening is shown by the two curves [52], for

(a) \( T_R = 1.28 \) \( t_e = 2.2 \)
(b) \( T_R = 1.30 \) \( t_e = 1.1 \).

surface stiffness \( \tilde{\alpha} \) (14). According to (9) and (11), the equilibrium size of facets is then orders of magnitude smaller than a typical dimension of the crystal. It cannot thus be directly measured, at least on the meniscus of figure 4.

If we extrapolate \( E_s \) down to 1.08 K, we find \( E_s \gtrsim 10^{-3} \) erg/cm\(^2\). In this case, we do not expect 2D nucleation to contribute to the growth below \( H \sim 10 \) mm. This is consistent with the observed absence of growth at \( T = 1.08 \) K in this range of \( H \) values. On the other hand, \( E_s \) extrapolates to zero at 1.23 K.

But, as the law (12) predicts that \( \frac{dE_s}{dT} \) vanishes at the roughening transition, this value is only a lower bound for \( T_R \). Indeed, when \( E_s/a \) is smaller than about \( 10^{-5} \) erg/cm\(^2\), \( R^* < \xi \) in the whole range of values of \( H \), and the surface grows as if it were rough. We can nevertheless try to estimate \( T_R \) by comparing the measured step energy to the theoretical prediction (26) \( E_s/a \sim kT_R/\tilde{\alpha} \), \( \tilde{\alpha} \) being calculated from equations (4)-(5). We find that the relatively slow measured variation of \( \log(E_s/a) \) in the temperature range 1.13 K-1.23 K implies that 1.23 K is at least 5-10 \% smaller than \( T_R \). This would explain our observation of facetted growth shapes up to 1.26 K, and also that the mobility \( V/H \) in figure 8 goes on increasing above 1.23 K. More precisely, qualitative agreement between the observed variation of \( \log(E_s/a) \) and the theory can be obtained if \( T_R \approx 1.28-1.30 \) K and \( t_e \) (see § 1.2) \( \sim 1-2 \) (15). For example, the full lines in figure 10 correspond to \( T_R = 1.28 \) K (1.30 K), and \( t_e = 2.2 \) (resp. 1.1). The corresponding asymptotic \( (t \to 0) \) behaviour of the step energy is \( E_s \sim \exp(-C/\sqrt{T}) \) with \( C = 1.5 \) (1.9) (16). This is quite reasonable, since, for example, in the \( \times y \) model, \( C \sim 1.5 \) [3], while in the six-vertex model, \( C \sim 2.1 \) [17]. However, a definite test of the expected theoretical behaviour would obviously require measurements of the step energy at lower and higher temperatures. To this end, the growth kinetics must be analysed at much higher and lower values of \( \frac{\Delta \mu}{k_B T} \) than those accessible by the technique here described. Never-theless, we shall now see that the validity of the present theory is strengthened by our measurements of the surface stiffness.

4. The surface stiffness of the (0001) orientation.

The principle of our experiment has been described in section 2. According to equation (14), the surface stiffness \( \tilde{\alpha}(0001) \) is given by \( \tilde{\alpha} = \frac{\Delta \rho g H R}{2} \). We measure the difference of levels \( H \) between the top of the meniscus (the \( c \)-axis of which is vertical) and the outer interface with a cathetometer. The uncertainty primarily comes from the position of the outer level, which appears as a thick dark line. It is systematic and about \( +0/ -0.1 \) mm (i.e. 3 \% for typical \( H \) values of 3 to 4 mm). In order to measure the curvature \( \frac{1}{R} \) in the \( c \)-direction, we first need to make a large picture of the meniscus. To this end, we use a parallel and horizontal light beam from a sodium lamp to make a magnified (x 20) image of the meniscus through an enlarger lens. Since the solid and liquid optical indices are very similar (\( \Delta n = n_s - n_l \sim 3 \times 10^{-3} \)), there is little diffusion by the meniscus. Its profile appears as a dark line (Fig. 11a) only because of refraction phenomena. The best definition is thus reached when the incident beam precisely coincides with the lens optical axis. After a further

\begin{align}
(14) & \text{ This order of magnitude explains why the spiral growth is here irrelevant. As we shall see in section 5, a typical growth rate due to dislocations is } V_d \approx V_e a/20 R^*, \text{ where } V_e \text{ is the step velocity. If we assume, as discussed in the text, that the velocity } V_{2D} \text{ due to 2D nucleation is } V_e \exp(-H^*/H), \text{ we indeed find that the ratio } V_d/V_{2D} \text{ is smaller than } 5 \% \text{ for any point in figure 9.}
\end{align}

For such high \( \xi^* \)'s, \( \xi \) is not given by equation (8). We have thus calculated its behaviour by an exact integration of equations (4)-(5). However, as these equations are themselves only valid for small \( \xi^* \)'s, the obtained behaviour of \( \xi \) is only approximate.

\begin{align}
(15) & \text{ We here use the relation } c = \frac{\pi}{2} \left( \frac{1}{t_e} + \frac{1}{2} \right)^{1/2} \text{ which is the generalization for large } t_e \text{ of the simpler relation (12) } c = \frac{\pi}{2} t_e^{-1/2} \text{ (valid only for } t_e \to 0) \text{ : it is easily derived from the exact integration of equations (4)-(5) [52].}
\end{align}
Fig. 11. — Enlarged views of two meniscii similar to that of figure 4. The hole diameter is 0.83 mm; a) for a vertical c-axis, at a temperature 1.25 K; b) for a vertical a-axis, at a temperature 0.95 K (the c-axis is horizontal and nearly in the plane of photograph). The curvature has a pronounced minimum near the c-axis (a) but not near the a-axis (b).

eNRiop expansion. In order to avoid this problem, we melt the crystal below the bottom of the box and grow it again, before performing a cycle at a new temperature. (We checked that, at a given temperature, the measured value of \( \bar{\alpha} \) does not change from one meniscus to another). This procedure thus allows us to study a meniscus free of visible defects up to 1.40 K.

The question is now to draw the surface stiffness from the crystal profile. A simple idea would be to describe this profile, near the c-axis and above the roughening temperature, by the expansion

\[
\alpha(x) = A(x - x_0)^2 + B(x - x_0)^4 + z_0
\]

where \( z \) is the coordinate along the c-axis and \((x_0, z_0)\) are the coordinates of the top of the meniscus. (In Andreev's theory [19], \( A \) should vanish at \( T_R \)). In fact, according to the theory described in § 1.2, the expression of \( \alpha(x) \) should also include more complicated terms, due to the non-analytical dependence of \( \bar{\alpha} \) on the angle \( \phi \) near the c-axis (i.e., near \( \phi = 0 \)). Nevertheless, the calculation shows that the theoretical surface stiffness \( \bar{\alpha} \) varies strongly with \( \phi \) only for very small angles and then tends to \( \alpha_0 \) very slowly. In the accessible range of \( \phi \) values — which is limited by our resolution to \( \phi \leq 0.05 \) rd, one can define a weakly \( \phi \)-dependent \( \bar{\alpha} \), so that the crystal shape should be properly accounted for, within our experimental resolution, by an expansion similar to (16). Here, however, \( 2A \) would not be the actual curvature at \( \phi = 0 \), but rather some angular average of the curvature for \( \phi \leq 0.05-0.1 \) rad. As for the \( Bx^4 \) term, it accounts for the \( \phi \) dependence of \( \bar{\alpha} \) for larger angles, to which the « crystalline » anisotropy (i.e. that not due to renormalization effects) may contribute. (Note that, because the hole has a small diameter, the effect of gravity is negligible in the angular windows used). If we assume an axy-symmetric meniscus near the c-axis, it is easy to show that

\[
\frac{1}{\tilde{\alpha}_1} \frac{\partial^2 \tilde{\alpha}_1}{\partial \phi_1^2} = \frac{1}{R} \frac{\partial^2 R}{\partial \phi_1^2} = 3(1 - B/A^3)
\]

where \( \tilde{\alpha}_1 \) is the surface stiffness in the plane of observation and \( \phi_1 \) is the polar angle in the same plane measured from the c-axis. Starting from a digitized profile, we determine \( x_0, z_0, A, B \) and the precise orientation of the c-axis by using a least-squares fit in an angular window of about \( \pm 0.3 \) rad with respect to the picture vertical direction. In order to test the validity of the expansion (16), we recalculate \( A, B, z_0 \), keeping the two other parameters fixed, in successive angular windows of \( \pm 0.25, \pm 0.20 \) and \( \pm 0.15 \) rad respectively.

Figure 12 presents the curvature versus temperature, as measured in the largest window. The curvature has been normalized by the theoretical jump (6),
The measured curvature $1/R$ of the $c$-orientation as a function of temperature. It is normalized by its theoretical value $\Delta P a^2/\pi k_B T_R$ at the roughening transition, so that the vertical unit is $\pi k_B T_R/2 \bar{a}(T) a^2$ ($T_R$ is taken as 1.28 K). The curvature $1/R$ is obtained by fitting the profile (Fig. 11a) with an equation of type $z = AX^2 + BX^4$. The fit is made in an angular range of $\pm 0.3$ rad with respect to the $c$-axis ($1/R = 2A$). $\Delta P$ is derived from the level difference. Different symbols refer to different values of $H$ (or $\Delta P$), $\bar{a}$: $2.9 < H < 3.1$; $+ 3.1$ to $3.3$; $\Box 3.3$ to $3.5$; $\Diamond 3.5$ to $3.7$; $\bigcirc 3.7$ to $3.9$, $\ast H > 3.9$ mm). Above $T_R = 1.28$ K, the normalized curvature is nearly unity. This indicates the validity of the present theory of the roughening transition. The behaviour below 1.14 K is discussed in the text.

The error bars on $T_R$ include both the uncertainty $\Delta T = 0.245 \pm 0.01 \pm 0.014/ + 0.007$ erg/cm$^2$ where the last term is the systematic error ($< 6\%$). This is in perfect agreement with the value determined for example at 1.20 K

$< \bar{a} > = 0.242 \pm 0.004 - 0.014/ + 0.007$ erg/cm$^2$

where the value is the systematic error ($< 6\%$). This is in perfect agreement with the value determined for example at 1.20 K

$\bar{a}(1.20\text{ K}) = 0.245 \pm 0.01 \pm 0.014/ + 0.007$ erg/cm$^2$

and slightly larger than the value $0.21 \pm 0.01$ simultaneously obtained in Moscow by a different method.
In terms of the normalized curvature $C$, we have

$$\langle C \rangle = 1.28 - 0.10/ + 0.14.$$  

The error here includes the precision on our temperature measurements (3%). The same analysis carried with the three smaller windows yields identical results, but with larger error bars. As an example, for the smallest window ($\pm 0.15$ rad), we find $\langle C \rangle = 1.24 - 0.14/ + 0.18$. The anisotropy at large angles is thus properly accounted for by the $B^4$ term in (16). Figure 13 shows the value of $B/A^3$, once averaged on the different height and in temperature intervals of 0.01 K (we checked that there is no significant dependence of $B/A^3$ on $H$). The relative error is much larger than it was for the curvature measurements, but our results remain consistent with a temperature independent value $B/A^3 = 9 \pm 2$. According to (17) this means that $\bar{z}$ is maximum in the c-direction, with an anisotropy

$$\frac{1}{\bar{z}} \frac{\partial^2 \bar{z}}{\partial \phi^2} = -24 \pm 6 \text{ rad}^{-2}.$$  

This fairly large anisotropy (19) clearly appears in the picture of figure 11a. The curvature at 30 degrees from the c-axis is obviously larger than in the vertical c-direction. Even at 0.2 rad from the c-axis, it is already 50% higher than on the c-axis. This corresponds to a surface tension $\bar{\sigma}(0.2 \text{ rad}) = 2/3 \bar{\sigma}(0) \simeq 0.16 \text{ erg/cm}^2$ while at 0.3 rad, $\bar{\sigma}$ is about 0.13 erg/cm$^2$. These two values are typical of the average surface tension of hcp $^4$He [11].

The normalized curvature is thus at least 10% to 15% larger than the theoretical value $C$ at the transition. However, this difference does not contradict the present theory of the transition. Indeed, we recall that we do not actually measure the surface stiffness $\bar{\sigma}(\phi = 0)$ but rather some angular average around $\phi = 0$, which is in principle lower than $\bar{\sigma}(\phi = 0)$. In order to evaluate the effect of this procedure, we plotted in figure 14 the theoretical temperature dependence of $C(\phi) = \pi \frac{k_B T R}{2 \bar{\sigma}(\phi) \sigma^2}$ for $\phi = 0, 0.1$ and 0.3 rad and $t_c = 1(20)$. The difference between these two functions is not significant: $\phi = 0$ and $\phi \neq 0$ appears quite appreciable. Consequently, our experimental points are consistent, within the errors bars, with $\bar{\sigma}(0.1 \text{ rad}, T)$ or $(0.3 \text{ rad}, T)$, while they are not with $\bar{\sigma}(0, T)$. Thus, our experimental procedure for determining $\bar{\sigma}$ can actually be responsible for the observed difference between our results and the theoretical behaviour of the surface stiffness. Taking also into account the approximate characters of the theoretical calculation of $\bar{\sigma}(\phi)$ (see note (19)) and of our analysis (21), we finally believe that the agreement

Fig. 14. — Comparison of our experimental results to the theoretical normalized curvature calculated for $T_R = 1.28$ K, $\phi = 0, 0.1$ and 0.3 rad and $t_c = 1$ (not that, for all the theoretical curves, the asymptotic value $\frac{\pi k_B T R}{2 \sqrt{2} \sigma^2} = 1 + \frac{t_c}{2} = 1.5$ is far from being reached at $T = 1.1$ $T_R$).

(19) As for $\xi(T)$ (see note (15)), we have calculated $\bar{\sigma}(\phi, T)$ by an exact integration of equations (4)-(5), without any approximation (like $t \ll 1$ or $t_c \ll 1$). But here also, the equations themselves are only approximative for $t_c \sim 1$.

(20) In particular, the deviation, at low temperature, of our measurements from the theoretical curves for $\bar{\sigma}(\phi, T)$ could be due to the existence of a highly curved region ($z \sim x^{3/2}$) near the facet edge, which is not accounted for by expansion (16). We can also note that the statistical error bars on $A$ in equation (16)(and hence on $C$) may be somewhat underestimated, since they assume the validity of expansion (16).
is satisfying, although a definite conclusion would of course require much more precise measurements of \( \bar{a}(0, T) \)\(^{(22)}\).

5. Study of the (1\overline{1}00) orientation.

In this section, we present our measurements of both the surface stiffness and the growth kinetics of the (1100) surface. In order to test the prediction (10) for the roughening temperature of the (1100) orientation, one must know the surface stiffness in the two principal sections of the (1100) surface which are respectively normal to the [1\overline{1}20] and [0001] axes. Indeed, there is only a two-fold symmetry around the [1100] axis and the two principal surface stiffness \( \bar{a}_1 \) and \( \bar{a}_2 \) can then be different. In this case, our experimental method, which we described in section 2, presents some difficulties. For a low enough level difference \( H \), that is for a nearly flat meniscus, the hole circularity implies \( R_1 = R_2 = (\bar{a}_1 + \bar{a}_2)/(\Delta \rho g H) \), so that we measure \( \bar{a}_1 = \bar{a}_2 = \frac{\Delta \rho g H}{2} \), and the measurements of \( R_1 \) would yield

\[
\bar{a}_1 = \frac{\Delta \rho g H}{2} \quad \text{(Eq. (9)).}
\]

We have studied a crystal with a horizontal a-orientation by the same method as in section 2. The only difference is that we used a smaller box (2 \times 2 millimeters). The hole diameter was the same (0.83 mm). Since the experiment has been carried in our dilution cryostat, which has only two sets of windows, only one curvature radius could be measured on a given crystal. We report here results on the surface tension \( a_1 \) in a section nearly normal to the [1\overline{1}20] axis (5 degrees off), which was got by the same analysis as in section 4. First, we find that the product \( R_1 H \) is constant with \( H \) between \( H = 3.9 \) mm and \( H = 4.7 \) mm, corresponding to values of the angle \( \theta \) between the meniscus edge and the horizontal plane between 0.7 and 1.1 rad. We are therefore in one of the two limits discussed above, so that we find \( (\bar{a}_1 + \bar{a}_2)/2 \) or \( \bar{a}_1 = 0.19 \pm 0.02 \) erg/cm\(^2\) (depending on which limit is realized). This must be compared to the value \( \bar{a}_2 \) of the surface stiffness in the section normal to the c-axis, which has been recently found equal to 0.25 erg/cm\(^2\) [41]. As for the surface stiffness anisotropy away from the a-axis, we find that, unlike for the c-orientation, the ratio \( B/A^3 \) of the coefficients in expansion (16) is much less than 1.

According to relation (17), we then have

\[
\frac{1}{a_1} \frac{\partial^2 \bar{a}_1}{\partial \phi^2} \approx 3.
\]

The surface stiffness is thus minimal in the (1100) direction, instead of maximum in the (0001) direction. This difference clearly appears in figure 11.

We can try to evaluate the roughening temperature of the (1100) orientation according to expression (10):

\[
k_B \, T_R = \frac{2}{\pi} a^2 (\bar{a}_1 + \bar{a}_2)^{1/2}. \quad \text{Here, } a \text{ is the periodicity of (1100) surfaces along the [1\overline{1}00] direction (} \bar{a} = 3.18 \text{ Å).}
\]

Note that, because the hcp lattice is not a Bravais lattice, \( a \) is different from the spacing between successive (1100) planes, which is either \( a/3 \) or \( 2a/3 \). We get for \( T_R \) the theoretical estimate (depending on whether the measured quantity is \( (\bar{a}_1 + \bar{a}_2)/2 = \bar{a}_1 \) or \( \bar{a}_1 \)) 0.90-1.05 K. It is in fair agreement with the estimation 0.95 K < \( T_R < 1.07 \) K which we shall draw from our study of growth kinetics. We remark that, if the a-orientation were (1120) instead of (1100), the theory would predict a much too low roughening temperature. Indeed, unlike the (1100) planes, the (1120) planes are equally spaced. Their periodicity is then about half that of (1100) planes (1.84 Å instead of 3.18 Å), which would give \( T_R \approx 0.3 \) K. We thus believe, as mentioned in the introduction, that the a-orientation is (1100) rather than (1120).

In order to test further the prediction (10), we could try to see whether it applies to other orientations. Unfortunately, the polar plot of \( \bar{a} \) is not precisely known enough for an accurate test. We only mention that (10) would yield the correct value for the (1101) roughening temperature [14] \((T_R \approx 0.35 \) K) if \( (\bar{a}_1 + \bar{a}_2)^{1/2} \approx 0.10 \) erg/cm\(^2\). This is a reasonable value. Surface tensions of the same order have been indeed measured in various orientations [11]. Let us also note that for the bcc \( ^4 \)He crystal, which is stable between 1.46 K and 1.77 K, \( \bar{a} \) is less than 0.12 erg/cm\(^2\). The highest roughening temperature, corresponding to the (110) planes, would then be 0.45 K. This explains why bcc crystals are rough [48].

We now come to the study of growth kinetics of the (1100) orientation. This study has been carried out by the same method as in section 3. The results, which are much more complex and irreproducible than for the c-facet near its roughening transition, are consistent with a dislocation mediated growth, far enough below \( T_R \). They also show a new phenomenon, which is an increase of the mobility \( (23) \) when the temperature decreases below 0.4 K.

Let us first describe how the relaxation takes place. In the first stage of growth, the small crystal inside the box is not in contact with the walls (Fig. 15a). When one of its lateral facets reaches a wall, the crystal often gets unstable and fills the whole box width, in a time

\( (22) \) Such measurements would allow to test the influence of quantum effects on the critical behaviour of \( \bar{a}(0, T) \). The theory treats the fluctuations in a classical way, while, if one extrapolates the ripplon dispersion relation up to \( \omega \approx 100 \) MHz, one can expect quantum effects for wavelengths smaller than \( \sim 50 \) Å at 1 K.

\( (23) \) Except above \( T_R \), we now use the term of mobility only in a qualitative sense. Indeed, we shall see that \( \bar{V} \) is generally not proportional to \( \Delta \mu \).
Fig. 15. — Growth of a meniscus with a horizontal $a$-facet at 0.16 K. The successive stages are obtained at a fixed outer level. The lateral faces are $c$-facets. (The box width is 2 mm.)
shorter than 1/50 s (which is the time interval between two video scans). The horizontal growth velocity involved in this process is thus larger than several centimeters per second, while the vertical velocity usually remains much smaller. During this phenomenon, the horizontal motion of the interface is likely due to the growth of rough orientations. Once the whole box width is filled, the facet size is much larger than before (Fig. 15d). The growth then goes on in a more regular way. Figure 16 shows the growth velocity $V$ as a function of the level difference $H$ for two different runs at 0.4 K. We first note that $V$ is about 10 microns per second for $H = 5$ mm. This is six orders of magnitude smaller than what would be the velocity of rough orientations for the same $\Delta \mu$ and temperature, which demonstrates the drastic difference between the growth of smooth and rough surfaces. This is of course consistent with the observed normal transmission (i.e. given by the Khalatnikov theory) of sound [49] and heat [50] across the facets. For the first run, an increase of $V$ follows the filling of the box (indicated by an arrow). Such a 2 to 4 fold increase is often observed but is not systematic. Indeed it does not happen for the second run, where the growth velocity before and after the contact is about the same. Therefore, the acceleration of growth in the first run is probably not due to the size increase of the horizontal facet (Fig. 15) which occurs for both runs. It is rather a consequence of some modification in the facet microscopic state, which could be due to stresses developed during the rapid filling of the box (for instance, this could create rows of dislocations, which likely give large growth rates [1]). Even besides this phenomenon, there are many other irreproducible features. For instance, although the relation $V(H)$ is about the same for $H < 3$ mm in curves 1 and 2 of figure 16, this is not the general rule. As a matter of fact, the typical velocities in this range of values of $H$ can vary by a factor of 10. Moreover, the curves in figure 16 are rather smooth. In other cases, the relation $V(H)$ is much more chaotic and can even be non monotonic. This could indicate that the vertical growth velocity $V$ may also depend on other facets than the horizontal one.

Although this complex behaviour makes difficult a quantitative study of the whole relation $V(H)$, some general conclusions can nevertheless be drawn. First, $a$-facets are observed to grow even at the lowest temperature reached (80 mK $< T_{th}/10$) for relatively small values of $\Delta \mu$ ($H \sim 2-5$ mm). Together with the irreproducibility of our results, this cannot be explained by 2D nucleation. At least below 0.8 K, the steps necessary for the growth are thus mainly supplied by screw dislocations. Since the number and relative arrangement of these dislocation vary from one experiment to another, and also with time in a given experiment (because the facet size evolves), this explains the complex behaviour observed. This growth process generally involves a threshold $[1] \Delta \mu_a = \frac{2}{\rho_s} (a d P)$ where $E_s$ and $a$ are the step energy and height and $d$ the distance between dislocations of opposite sign ($\Delta \mu_a$ is the minimal $\Delta \mu$ necessary to free the steps attached between the dislocations). The final level $H_w$ is the larger value of $(\rho_s/\rho_l) \Delta \mu_l$ and $H_o$, the capillary depression in the 2 x 2 millimeters box. Between 0.4 K and 1 K, $H_w$ is constant and about 1.2 mm. When the temperature decreases below 0.3 K, $H_w$ increases on an average from 1.2 to 2 millimeters, with some dispersion from one experiment to another. This means that the capillary depression is $H_o = 1.2$ mm ($24$) and that the threshold $\Delta \mu_a(T)$ is given by $(\rho_s/\rho_l) g H_o(T)$ only below 0.4 K. Anticipating the following estimation of the step energy, $E_s[a \sim 2 \times 10^{-2}$ erg/cm$^2$ at low temperature ($T = 0.15$ K), we deduce from the values of $\Delta \mu_a$, a typical size of active dislocations pairs $d = 100$ microns. This is not very small compared to the size of the growing facet, so that finite size effects may also be important in our experiments.

We now estimate the step energy from the following experiment. We first fill the box with a crystal at the temperature 0.9 K, i.e. near $T_{th}$. The inner level is then lower than the outer one by the capillary depression $H_o$. When we decrease the temperature at a constant mass in the cell, the outer level remains fixed, because the thermal expansions of both liquid and solid phases are negligible below 0.9 K. As $H_o$ likely does not depend on temperature, the facet does not have to move vertically when the temperature changes. Thus,

![Fig. 16. — Two typical growth characteristics for a horizontal $a$-facet at 0.4 K. The arrows indicate the contact of the growing crystal with the walls of the box. The irreproducibility suggests that the dominant mechanism is spiral growth.](image.png)
the interface equilibrium shape is reached by the motion of rough orientations only. The true equilibrium size of the facet can then be measured without any problem of threshold. We find from equation (11) \( E_g \approx 1.5 \times 10^{-2} \text{ erg/cm}^2 \) at 0.4 K and \( 2 \times 10^{-2} \text{ erg/cm}^2 \) at 0.15 K. Let us note that this method cannot be applied to the c-facet, because the thermal expansion around \( T_R \approx 1.28 \text{ K} \) is important.

Finally, we come to the study of the growth kinetics as a function of temperature. Since, at a given temperature, the relation \( V(H) \) is not reproducible, we decided to study only the growth velocity \( V \) at the beginning of growth. The measurements are performed for a given level difference \( H \approx 5 \text{ mm} \) and before the first contact with the walls takes place. This strongly reduces the irreproducibility. Figure 17 shows that \( V \) increases both at high and low temperatures. Because the level difference in this experiment is at least twice as large as the above mentioned thresholds, the qualitative temperature dependence of the growth velocity \( V \) should be the same as for a single dislocation. In this case, the step issued from the dislocation is wound in a rotating spiral. The growth velocity \( V_g \) is \( V_g \approx \frac{a}{l} \), where \( a \) and \( V_g \) are the step height and velocity, and \( l \) is the distance between successive turns : \( l \) is about twenty times [1, 51] the critical radius nucleus, so that \( V_g \approx a^2 \rho_0 \Delta \mu/2 E_s \). Since the step energy increases when the temperature decreases, the step velocity \( V_g \) (and hence the step mobility \( k_s = V_g/\Delta \mu \) ) must behave in the same way in order to explain our low temperature results. This increase of the step mobility as \( T \) tends to zero shows that, as the rough surface mobility, the step mobility is limited by some thermal excitations (phonons or surface excitations). Since we do not expect this phenomenon to depend on the facet orientation, the mobility of c-facets should also increase at low temperatures. This is qualitatively observed. For a level difference \( H \approx 5 \text{ mm} \), a c-facet grows much more easily at 0.1 K than at 0.5 K, while it does not grow at all at 1 K.

We can go further if we assume that the facet velocity \( V \) has exactly the temperature dependence of \( V_g \), i.e. \( V \approx k_s/E_s \). Our measurements then indicate that \( 1/k_s \approx T^{-3} \), within an additive constant, more like \( T^2 \) or \( T^4 \), which is the measured law for a macroscopic rough interface [11]. Indeed, a \( T^4 \) or \( T^3 \) variation would imply a much more rapid decrease of \( V \) than observed, except if the step energy abruptly drops between 0.2 and 0.4 K (for a \( T^4 \) variation, \( E_s \) would be divided by 20 between 0.2 and 0.4 K). This is neither reasonable, nor in agreement with the above determination of the step energy. On the contrary, a \( T^2 \) law would be consistent with our measurements. However, a more quantitative analysis is difficult. For instance, the temperature dependence of \( V \) could also be affected by size effects. Indeed, for \( E_s/a = 2 \times 10^{-2} \text{ erg/cm}^2 \) and \( H = 5 \text{ mm} \), the distance between the successive turns of a rotating spiral is about 0.4 mm, i.e. of the order of the facet size.

On the other hand, when the roughening transition is approached, the step energy tends to zero. This enhances both the growth velocity \( V_g \) due to a single dislocation and the probability of 2D nucleation. These two mechanisms add together to cause the observed increase of \( V \) close to \( T_R \). At an intermediate temperature, the competition of the temperature dependences of the step mobility and energy leads to the observed minimum of \( V \) between 0.4 and 0.6 K. Finally, we estimate the value of \( T_R \) as follows. At 1.07 K, we observe that \( V \) is linearly related to \( H \) down to the capillary height (26). On the contrary, at 0.95 K, \( V(H) \) depends linearly on \( H \) only at high values of \( H \), with a slope twice smaller than at 1.07 K. Lastly, the mobility at 1.07 K is \( k_s \approx 5 \times 10^{-3} \text{ s/cm} \), which is equal to the extrapolated [47] mobility of

\footnote{The maximal observed values are about 100 \( \mu \text{s} / \text{s} \) at 0.1 K and several \( \mu \text{s} / \text{s} \) at 0.5 K.}

\footnote{Let us recall that, at 1 K, the c-facets do not grow in our experimental conditions. We then stay in the situation of figure 15c and never get that of figure 15d. In this case, \( H_0 \) is about 2.5 mm.}
rough orientations at this temperature. We thus believe that the roughening temperature lies in the interval 0.95 K-1.07 K. (Strictly speaking, as in section 3, this is only a lower evaluation for $T_R$.)

6. Conclusion.

Let us review our results and their connection to the theory of the roughening transition. As far as we know, our study of the growth kinetics is the first quantitative evidence of the continuous evolution from the smooth to the rough dynamic behaviour, when crossing a roughening temperature. It yields values for $T_R$, and $T_{R'}$, which are found slightly higher than previously thought. Near the roughening transition, the increase of the interface mobility is mainly due to 2D nucleation. This allows us to measure the temperature dependence of the important parameter as the roughening transition is approached, the facet mobility increases, due to the continuous vanishing of the step energy, which enhances both the activity of spiral growth and the probability of 2D nucleation.

For both the (0001) and (1100) surfaces, the crystal curvature remains finite at their respective roughening temperatures $T_R$. Moreover, the theoretical relation between $T_R$ and the surface stiffness $\tilde{\alpha}$, $k_B T_R = 2/\pi \tilde{\alpha}^2$, is fairly verified in both cases. In the case of the (0001) surface, for which the measurements are the more precise, the surface stiffness is slightly less (10% at least) than its theoretical value. This can be due to our experimental method, which only yields an average of the surface stiffness in a finite angular range. A definite test of the theory would require to measure the surface stiffness in an angular range around the c-axis smaller than, say, $10^{-3}$ radian. However, this seems difficult to achieve with the presently available techniques. In any case, we stress that both our results on the step energy and the surface stiffness are consistent with a rather strong coupling of the (0001) interface to the crystal structure.

Lastly, we also report measurements of the (1100) facet growth kinetics far below its roughening transition. The steps are then supplied by the screw dislocations (spiral growth). We find that the mobility of facets presents a minimum as a function of temperature. Starting from low temperatures (0.1 K), it first decreases, which shows that the mobility of microscopic steps is limited by their interaction with some thermal excitations. Then, at higher temperature, as the roughening transition is approached, the facet mobility increases, due to the continuous vanishing of the step energy, which enhances both the activity of spiral growth and the probability of 2D nucleation.

References


