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## From vicinal to rough crystal surfaces

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**Résumé.** — On s'attend généralement à ce que les propriétés d'une surface vicinale ne soient plus contrôlées par l'existence des marches lorsque celles-ci se recouvrent, donc lorsque leur distance mutuelle devient inférieure à leur largeur. En reprenant la théorie de la transition rugueuse élaborée par Nozières et Gallet [1], nous montrons que, pour des surfaces faiblement couplées au réseau cristallin, ce recouvrement doit se produire pour des distances nettement plus grandes que la largeur (telle qu'elle est habituellement définie). Notre prédiction est confirmée par l'analyse des différentes mesures de la variation angulaire de la rigidité de surface des cristaux d'hélium réalisées par Wolf *et al.* [2], Andreeva *et al.* [3] and Babkin *et al.* [4]. Il s'ensuit que l'étude de l'interaction entre marches cristallines doit être effectuée sur des surfaces vicinales d'inclinaison beaucoup plus faible qu'on ne le croyait jusqu'à présent. Cet article est aussi une occasion pour revenir sur les liens qui existent entre largeur des marches et longueur de corrélation des surfaces lisses, ainsi que sur le traitement des différents effets de taille finie qui apparaissent dans les problèmes de rugosité. Nous reconsidérons enfin comment l'hypothèse d'un couplage faible s'applique au cas des cristaux d'hélium.

**Abstract.** — One generally expects the properties of a vicinal surface to be independent of the existence of steps as soon as these steps overlap, i.e. when their mutual distance is smaller than their width. By using the roughening theory by Nozières and Gallet [1], we show that, at least for surfaces weakly coupled to the lattice, this overlap occurs for distances significantly larger than the commonly defined width. Our prediction is supported by an analysis of the various measurements of the angular variation of the surface stiffness of helium crystals, which were performed by Wolf *et al.* [2], Andreeva *et al.* [3] and Babkin *et al.* [4]. As a consequence, the interaction between crystal steps should be studied on vicinal surfaces with a much smaller tilt angle than previously thought. This article is also an opportunity to return to the relation between the step width and the correlation length on smooth surfaces, as well as to the treatment of the various finite size effects which occur in the problem of roughening. We finally reconsider how the weak coupling hypothesis applies to the case of helium crystals.

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Consider a crystal surface whose normal  $\langle n \rangle$  is tilted by a small angle  $\phi$  with respect to  $\langle n_0 \rangle$ , one of the high symmetry axes. If  $\phi$  is small enough, the surface is called vicinal and it can be described as a set of terraces limited by steps, which are well defined fluctuating lines with a certain free energy  $\beta$ , height  $a$ , width  $w$ , also an average spacing  $d = a/\tan \phi$  and some (presumably repulsive) interactions. However, as the tilt angle increases above some critical value  $\phi_c$ , this description should break down because steps overlap and terraces disappear. The main purpose of this article is to predict the value of  $\phi_c$ .

The analysis below applies to the temperature domain  $T_{R_n} < T < T_{R_0}$  between the respective roughening temperatures of the  $(n)$  and  $(n_0)$  surfaces (if  $\phi$  is small,  $T_{R_n}$  is very small compared to  $T_{R_0}$ ). As a consequence, steps exist and are free to fluctuate, they are not registered on the lattice, we do not consider the possible roughening transition of the vicinal surfaces  $(n)$  and their surface stiffness  $\gamma(\phi)$  remains finite.

## 1. Introduction.

Andreeva *et al.* [3] recently presented measurements of the surface stiffness anisotropy of hcp helium 4 crystals. Of particular interest are the data corresponding to surface orientations close to the basal plane. As a consequence of a  $1/d^2$  repulsion between steps ( $d$  is the step-step average distance), one generally expects the surface stiffness of vicinal surfaces to vanish linearly when their tilt angle  $\phi$  tends to zero. Instead of this, Andreeva observed a rise. Some authors tried to interpret this surprising result in terms of step-step interactions with a longer range than  $1/d^2$  [5]. Other authors [6] considered these results as totally inconsistent with the commonly accepted theory of roughening and with the interpretation of the experiments of Gallet *et al.* [7]. Our interpretation is different : we think that their measurements are done at tilt angles which are not small enough to show a stepped character. We also find a very good agreement between the existing experimental results [2-7], and the theory of Nozières and Gallet [1] (if correctly used).

A crossover should exist between two different regimes. At small tilt angle, steps should be well separated, i.e. non-overlapping or non-degenerate, and weakly interacting. Only then should the crystal surface be truly vicinal, which means that it is very anisotropic and that it can be described in terms of terraces, steps and step-step interactions. At larger angle, the steps are degenerate because they overlap and the surface becomes really rough and nearly isotropic. The change between the two regimes is not a true phase transition, more a gradual crossover as a function of angle around a critical value  $\phi_c$ . Since the step width depends on temperature, the angle  $\phi_c$  should also be temperature dependent.

From dimensional arguments, one might expect this crossover to occur when the step width  $w$  is comparable to the interstep distance  $d = a/\tan \phi$  ( $a$  is the lattice periodicity or step height since we only consider single steps here, for simplicity). This would mean that  $\phi_c$  is of order 1 radian since, at low enough temperatures, one generally expects the step width  $w$  to be of order  $a$ . To predict a transition at such a large angle would not have much interest, since the angular distance between different facets is smaller than 1 radian in the same low temperature situation.

A first problem actually arises in the definition of the step width. In this article, we mostly restricted ourselves to crystal surfaces which are weakly coupled to the underlying lattice. This is because a good general theory [1] exists for such surfaces, but mainly because it applies to the  $c$  faces of helium 4 crystals which are under extensive and precise study. The coupling strength is related to the width of steps in the zero temperature limit (in the absence of thermal fluctuations) : the weaker the coupling, the broader the steps. One generally expects a weak coupling for liquid-solid interfaces in systems with weak interactions or for high Miller index

orientations, and probably a strong coupling for the simple faces of metal-vacuum interfaces. A more precise definition and discussion of the weak coupling situation is given at the end of this article. At this stage, it is enough to realize that, in Nozières' theory [1], the step profile  $z(x)$  is optimized and found to be

$$z(x) = \frac{2a}{\pi} \tan^{-1} e^{(v/\xi)} \quad (1)$$

One is thus tempted to consider  $\xi$  as the step width ( $x$  is a horizontal coordinate). However the function  $z(x)$  is such that the distance over which the height  $z$  varies from  $0.1a$  to  $0.9a$  is about  $4\xi$ , and we propose  $w = 4\xi$  as a more physical, although somewhat arbitrary, definition of the step width. Also note that the quantity  $\xi$  is not easy to calculate nor to measure as explained in the appendix.

By using the part of Nozières' theory which is devoted to tilted surfaces, we came to the conclusion that the vicinal or stepped character only occurs below about  $\phi_c \approx a/12\xi \approx a/3w$ , rather than  $a/\xi$  as suggested in reference [1], so that this critical angle is smaller by one order of magnitude than previously thought. Our prediction of a crossover from vicinal (or stepped) to rough behavior becomes meaningful. It should be observable, at least for solid-liquid interfaces where the coupling of the crystal surface to the lattice is small enough and especially in helium (around  $2.5^\circ$  at low enough temperature).

Since the parameters of Nozières' theory were adjusted by Gallet *et al.* [7] to fit three different series of measurements on the roughening transition of c facets on helium 4 crystals, it has been possible to compare our predictions with some preliminary results from the experiments of Andreeva *et al.* [3], and also from earlier experiments by Wolf *et al.* [2] and by Babkin *et al.* [4]. We found a good agreement. It means that Nozière's theory is further supported by these two or three independent series of experiments, contrary to what has been published elsewhere [6]. Babkin's data actually appear to us to be the most precise present evidence for the universal curvature of crystals at the roughening transition.

To our knowledge, the crossover from well separated to overlapping steps has not yet been observed. It seems particularly important to observe it if one wants to measure the interactions between steps. Several authors have tried to measure these interactions [8-12], but we think that no definite proof has yet been obtained of a better description by one particular law (a  $1/d^2$  repulsion ?) than by another one ( $1/d$  ?). We also consider this rather open question here when describing what should be the angular variation of the surface stiffness of vicinal crystal surfaces in the limit of very small tilt angle.

Finally, the problem of vicinal surfaces in helium gives us an opportunity to reconsider the whole problem of the use of Nozières' renormalization theory of roughening. We slightly correct some little mistakes in the previous work of Gallet *et al.* [7]. We also improve the treatment of finite size effects and reconsider the validity of the weak coupling hypothesis.

## 2. The roughening theory by Nozières and Gallet [1].

Nozières first considered simple surfaces, with a normal along  $\langle n_0 \rangle$ , one of the high symmetry crystal axes. He wrote the following Hamiltonian for a surface deformation :

$$H = \frac{1}{2} \gamma (\partial z / \partial x)^2 + V \cos (2 \pi z / a) \quad (2)$$

where  $\gamma$  is the surface stiffness and  $V$  is an effective lattice potential (the surface energy oscillates with equally spaced minima). Nozières treated equation (2) by a renormalization

technique and obtained the two following differential equations

$$\frac{dU}{d\varepsilon} = (2 - n) U \quad (3)$$

$$\frac{d\gamma}{d\varepsilon} = 2 \pi^4 A(n) \frac{U^2}{\gamma a^4}, \quad (4)$$

where  $n = (\pi k_B T / \gamma a^2)$ , and where the coefficient  $A(n)$  slowly varies around  $A(2) = 0.4$ . These two equations describe the coupled evolution of the lattice potential  $U = V/\Lambda^2$  and the surface stiffness  $\gamma$  as a function of the scale  $\Lambda$  at which fluctuations are averaged ( $\Lambda$  is homogeneous to a wavevector and  $\varepsilon = -\log(\Lambda/\Lambda_0)$ ). These very important equations are also considered as parametric equations for the renormalization trajectories in the  $(U, \gamma)$  plane. The physical meaning of these trajectories is the following : starting from initial conditions corresponding to the microscopic scale  $\Lambda_0$ , one progressively averages on fluctuations with larger and larger wavelength by increasing  $\varepsilon$ . The lattice potential and the surface stiffness evolve towards their macroscopic values in a way which crucially depends on temperature. At the roughening temperature  $T_R$ , the trajectory goes to a fixed point  $n = 2$ , so that Nozières obtains the universal relation of roughening

$$k_B T_R = (2/\pi) \gamma_R a^2 \quad (5)$$

As compared to other similar ones, Nozières' theory has a remarkable advantage for experimentalists ; it explicitly contains three adjustable parameters :

- the roughening temperature  $T_R$ ,
- the microscopic size  $L_0$ , or the cutoff wavevector  $\Lambda_0 = 1/L_0$ ,
- the strength of the coupling to the lattice  $t_c = [2 \pi^2(A(2))^{1/2} U_0]/(\gamma_0 a^2)$ ,

where the subscript 0 is used for microscopic, unrenormalized values.

Nozières also developed a theory for dynamic roughening. By this we mean the evolution of the growth rate around the temperature  $T_R$  where thermal fluctuations drive the surface from smooth to rough. We also mean the roughening induced by the growth around  $T_R$ . But it is different from a second type of dynamic roughening, which was considered by Kardar *et al.* [13]. In order to calculate the critical behaviour of the growth rate, Nozières applied the same renormalization method to the Langevin equation

$$\rho_c k^{-1} \partial h / \partial t = \rho_c \delta \mu + \gamma \Delta h - (2 \pi V/a) \sin(2 \pi z/a) + R(z, t) \quad (6)$$

where  $k$  is the growth rate or surface mobility,  $\rho_c$  is the crystal density,  $\delta \mu$  is the difference in chemical potential between the crystal and the liquid, i.e. the force driving the growth, and  $R$  is a random force describing the thermal fluctuations. This Langevin equation describes the motion of the crystal surface in the presence of an applied force, a capillary pressure, a lattice potential and thermal noise. Nozières arrived at three renormalization equations, two of which are very close to equations (3) and (4) while the third one describes the renormalization of the growth rate  $k$  :

$$\frac{dk}{d\varepsilon} = -8 \pi^4 B(n) k \frac{U^2}{\gamma^2 a^4} \quad (7)$$

where  $B(n)$  is a new coefficient reaching its maximum value 0.25 for  $n = 2$ .

The three parameters  $T_R$ ,  $\Lambda_0$  and  $t_c$  were fitted by Gallet *et al.* [7]. The step energy below the roughening transition, the average curvature around the  $c$  orientation and the critical variation of the growth rate for various departures from equilibrium all showed a good agreement with Nozières' predictions using the same set of values for  $T_R$ ,  $\Lambda_0$  and  $t_c$ .

In addition to the above treatment of equations (2) and (6), Nozières' theory contains another part, which describes tilted surfaces.

Nozières considers a tilted surface as a set of terraces with size  $d$ , the same average distance between steps as above. He then realizes that the fluctuations are limited to wavelengths smaller than about  $d$ , so that the properties of tilted surfaces are obtained from those of non-tilted surface if the renormalization procedure is stopped at a right size of order  $d$ . This looks simple but needs a correct estimate of this long wavelength cutoff, or exact finite size effect. In order to do it, he introduced the modified Hamiltonian

$$H = \frac{1}{2} \gamma (dz/dx)^2 + V \cos [2 \pi (z - \phi x)/a] . \tag{8}$$

The same renormalization technique was applied, with similar results and a main difference : a new coefficient  $A^{**}(n, C)$  replaces  $A(n)$ , which now depends on the scale  $\Lambda$  and on the angle  $\phi$  through the quantity  $C = \phi/a\Lambda$ . As shown in figure 1, we calculated numerically the quantity  $A^{**}(n, C)$  from the rather complicated analytical expressions

$$A^{**}(n, C) = \int_0^\infty r^3 dr \int_0^\infty \frac{dx}{x} e^{-\frac{1}{4}x} J_0(r) e^{-r^2 x} e^{-2nD(r, \nu)} J_1'(2 \pi Cr) \tag{9}$$

$$D(r, x) = \int_0^r \frac{du}{u} [1 - J_0(u) e^{-u^2 x}] \tag{10}$$

where  $J_0$  and  $J_1$  are Bessel functions (the ' indicating a first derivative). This figure shows that the new coefficient  $A^{**}(n, C)$  vanishes around  $C = 1/6$ . Consequently, according to this model, the renormalization stops at a maximum scale  $1/\Lambda_{\max}$  which is about equal to one sixth of  $a/\phi = d$ , the interstep distance. Furthermore, as explained in the appendix, the length

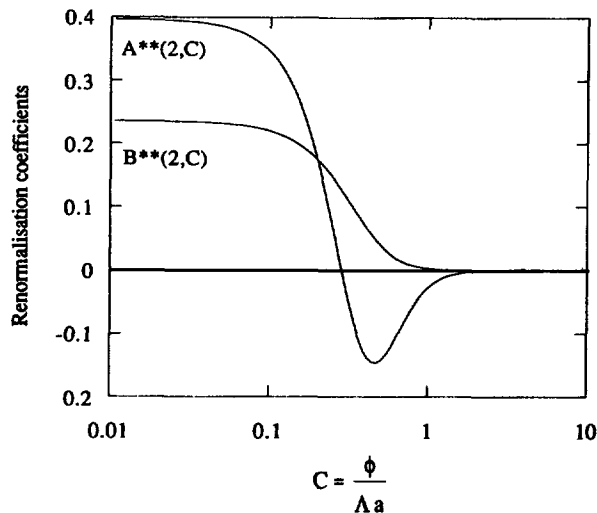


Fig. 1. — In the renormalization equations of Nozières' theory of roughening, the numerical coefficients are modified if the surface is tilted with respect to one of the main lattice planes. The modified coefficient  $A^{**}$  concerns the renormalization of the surface stiffness and vanishes around  $C = \phi/\Lambda a = 1/6$ . It means that the surface does not feel the effect of the lattice if the angle is larger than  $\Lambda a/6$ . ( $\Lambda$  is the inverse scale at which the fluctuations are averaged, it is inversely proportional to the step width). As a consequence, steps on a vicinal surface are well separated and non-degenerate only if their mutual distance is at least three times their width. The coefficient  $B^{**}$  concerns the growth rate of tilted surfaces and vanishes at a somewhat larger angle.

$\xi$  is about half the maximum scale  $1/\Lambda_{\max}$ . We thus arrive at the announced result : tilted surfaces can be treated as sets of terraces separated by well defined steps only if  $\xi$  is smaller than about  $d/12$  (i.e. if the step width  $w$  is smaller than about  $d/3$ ).

So, what happens when the tilt angle  $\phi$  decreases ?

If  $\phi > \phi_{c0} \approx a/12 \xi_0$ , the renormalization does not even start as  $1/\Lambda_0$  is larger than  $d/6$  and the surface stiffness  $\gamma$  has essentially the microscopic value  $\gamma_0$ .

In the range  $\phi_{c0} > \phi > \phi_c \approx a/12 \xi$ , renormalization starts at the scale  $1/\Lambda_0$  and stops because  $A^{**}(n, \Lambda)$  vanishes at a scale  $1/\Lambda_{\max}$  which depends on  $d$ . In this angular range, and close enough to  $T_R$ ,  $\gamma$  increases above the unrenormalized value  $\gamma_0$ , but steps are still rather close to each other and the terraces are not wide enough to be well defined smooth surfaces.

On the contrary, below  $\phi_c \approx a/12 \xi$ , the renormalization stops because the effect of the lattice has fully developed, terraces and steps are well defined, and surface fluctuations are killed by the effect of a large potential.

Of course, these numbers 3 or 12 are approximate and presumably model dependent, and our reasoning is not yet strong enough to show that the step overlap occurs around  $\phi_c = a/12 \xi$ . The evidence comes from the comparison with experiments, including data obtained in Moscow by Babkin *et al.* [4] and by Andreeva *et al.* [3]. In doing this, we wish to present the interpretation of our old curvature measurements [2, 7] in a slightly different way. As we shall see, the three parameters  $T_R$ ,  $\Lambda_0$  and  $t_c$  will not be significantly modified, the agreement will be extended to other independent measurements, and the whole analysis supports the determination of  $\phi_c$ .

### 3. Comparison with experiments in helium 4.

Wolf, Gallet *et al.* [2, 7] have measured three quantities : the step energy  $\beta$  below the roughening transition, the critical variation of the growth rate  $k$  and the average curvature of crystals.

The last measurement has been made in an angular window of 0.15 to 0.3 rad around the  $c$  direction. From a precise knowledge of the pressure at which thermodynamic equilibrium was achieved between the crystal and superfluid helium, they obtained the value  $\gamma = 0.245 \text{ erg/cm}^2$ . As shown by figures 2 and 3, good fits of the data concerning the step

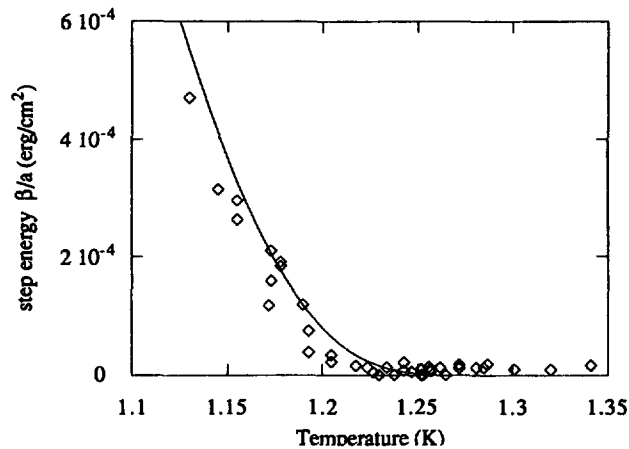


Fig. 2. — A new comparison between the step free energy as measured by Gallet *et al.* [7] and as calculated from Nozières' theory after readjustment of its three parameters ( $T_R = 1.30 \text{ K}$ ,  $t_c = 0.58$ ,  $\Lambda_0 = 0.25/a$ ).

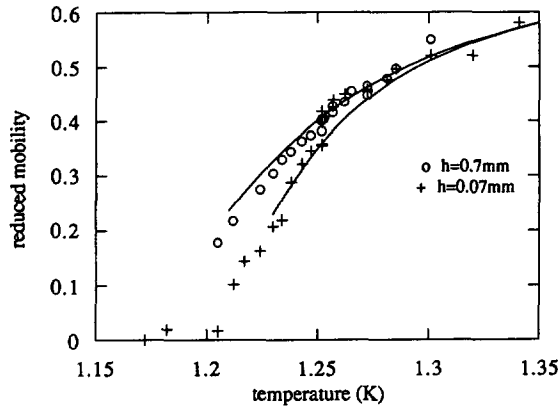


Fig. 3. — A new comparison of the growth rate of *c* surfaces as measured by Gallet *et al.* [7] and as calculated from Nozières' theory with the same choice of parameters as for the step energy (Fig. 2).

energy  $\beta$  and the growth rate  $k$  are obtained with  $\Lambda_0 \approx 0.25/a$ , so that the angle  $\phi_{c0}$  should be about  $0.04 \text{ rad} = 2.5^\circ$ . This means that Wolf, Gallet *et al.* could not measure the critical variation of  $\gamma$ , which only shows up in a very small angular range. However their measurement is still very useful : it gives the unrenormalized value of the surface stiffness

$$\gamma_0 = 0.245 \text{ erg/cm}^2 \tag{11}$$

Using this value and the following relation demonstrated by Nozières :

$$\gamma_R = \gamma(T = T_R, \phi = 0) = \gamma_0(1 + t_c/2), \tag{12}$$

we adjusted  $T_R$ ,  $t_c$  and  $\Lambda_0$  to obtain the curves shown in figures 3 and 4. One reason we made these fits again is that we found an unfortunate numerical mistake in one of Gallet's fitting

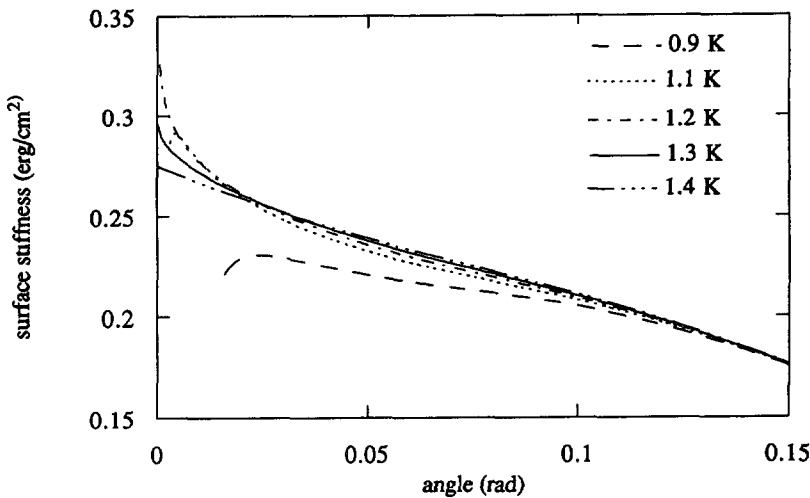


Fig. 4. — The angular variation of the surface stiffness of helium crystal surfaces. The angle  $\phi$  is the tilt with respect to the *c* or basal planes of the hcp structure. The successive curves correspond to different temperatures in the vicinity of  $T_R = 1.3 \text{ K}$ . The calculation includes the non-singular anisotropy of the crystal and the renormalization of the surface stiffness by the lattice potential.



programs. Our new values for the adjustable parameters are

$$T_R = 1.30 \pm 0.01 \text{ K} \quad (13)$$

$$t_c = 0.58 \pm 0.02 \quad (14)$$

$$A_0 = (0.25 \pm 0.04)/a \quad (15)$$

with  $a = 2.99 \text{ \AA}$ . These values are not significantly different from what was published in 1987 (1.28 K, 0.63,  $1.16/a$ ). The following other values can be deduced. At  $T_R$  and  $\phi = 0$ , the universal value of the surface stiffness is

$$\gamma_R = 0.315 \text{ erg/cm}^2 \quad (16)$$

in agreement with equation (5). At  $T = 0$ , we find a minimum step width

$$w_0 = 4 \xi_0 \approx (2/A_0) (2 \sqrt{A(2)/t_c})^{1/2} \approx 12 a, \quad (17)$$

a rather large value which partly explains why steps become well separated on helium crystals only below the small angle  $\phi_{c0} = aA_0/6 \approx 0.04 \text{ rad} = 2.5^\circ$  (in the low temperature limit).

We now turn to the angular dependence of the surface stiffness  $\gamma$ , in order to interpret the data obtained in Moscow [3, 4]. As explained in part 2, Nozières' theory can be used to calculate this angular dependence above the roughening transition, more precisely when steps overlap. It is valid as long as the correlation length is only limited by the system size. This is true above or at  $T_R$  where  $\xi$  would be infinite for an infinite crystal. It is also true below  $T_R$ , if  $\phi > a/12 \xi$ , on the large angle side of the crossover to vicinal behavior. It can thus be used to calculate  $\gamma(\phi)$ , not only at all angles above  $T_R$  but also below  $T_R$  for angles larger than  $\phi_c$ . This is what we tried, by integrating the renormalization trajectories with the new coefficient  $A^{**}(n, C)$ . In a preliminary publication [14], we briefly mentioned the results of a simplified calculation where equation (4) was kept with the coefficient  $A(n)$  but the renormalization was abruptly stopped at  $L_{\max} = 1/\Lambda_{\max} = a/6 \phi$ . These early results were not significantly different from the more rigorous calculation presented here and shown in figure 4.

Figure 4 calls for the following comments. We used the parameters which we previously adjusted to the case of  $c$  facets in helium 4. At  $T_R$  and when  $\phi$  tends to zero,  $\gamma(\phi)$  tends to the universal value  $\gamma_R$  with a singularity which was shown to be logarithmic by Nozières [1]. Close enough to  $T_R$ , the critical angular variation of  $\gamma$  corresponds to an increase of the stiffness when the surface feels the lattice (at small angle), and it only occurs below the angle  $\phi_{c0}$  at which the microscopic scale  $1/\Lambda_0$  is about one sixth of the interstep distance  $a/\phi$ . The crossover to stepped behavior should occur at  $\phi_{c0}$  only if  $T = 0$ , but, if  $T$  is intermediate between 0 and  $T_R$  (where the steps themselves vanish), the crossover occurs at the smaller angle  $\phi_c = a\Lambda/6 \approx a/12 \xi$ . The critical angle  $\phi_c$  vanishes at  $T_R$ . Above  $\phi_{c0}$ , the surface stiffness has the unrenormalized value  $\gamma_0(\phi)$  whatever the temperature.

Above  $T_R$ , and still below  $\phi_c$ , the surface stiffness increases up to values which are smaller than the universal value  $\gamma_R$ , as consequence of the well known « square root cusp » which appears in a  $(\gamma, T)$  plot.

An additional difficulty in the analysis of the experimental results comes from the existence of a large angle variation of  $\gamma$ , which reflects the large angle anisotropy of the crystal and has nothing to do with the critical phenomena under present consideration. In the range  $0 < \phi < 0.3 \text{ rad}$ , Wolf, Gallet *et al.* found this non-singular variation of the surface stiffness to be well described by

$$\gamma_0(\phi) = 0.245(1 - 12 \phi^2) \text{ erg/cm}^2 \quad (18)$$

The last measurements by Andreeva *et al.* [3] agree very well with the above formula. This large angle anisotropy was included in the calculation shown in figure 4.

In figure 5, we show a comparison with experiments. The dotted line corresponds to the

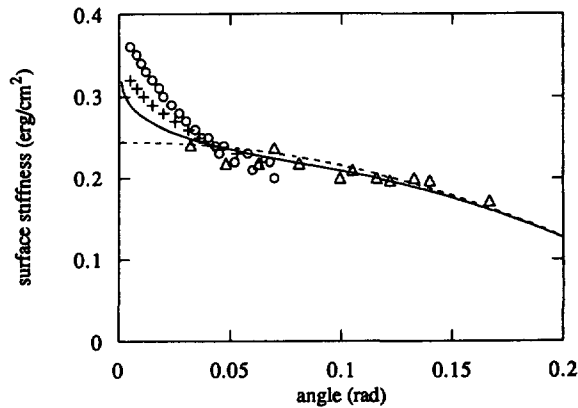


Fig. 5. — A comparison of Nozières' theory (solid line) adjusted on the experiments of Gallet *et al.* [7] with the experimental measurements around 0.4 K by Andreeva *et al.* (triangles) [3] and at 1.2 K by Babkin *et al.* (crosses and circles) [4]. The good agreement confirms that steps should become non-degenerate at low temperature only below 0.04 rad where an inflexion indicates the limit of the critical angular region. The dotted line corresponds to equation (18) in the text, i.e. to the large angle, non-singular variation of the surface stiffness. The solid line is calculated at 1.2 K in order to compare with the points by Babkin, which were taken at this temperature and are the only ones inside the critical angular domain where renormalization effects, consequently temperature variations, occur. Andreeva's data correspond to angles outside this domain and consequently agree both with theory and other experiments at higher temperatures.

non-singular part only, as described by equation (18). The solid line corresponds to  $T = 1.2$  K, the temperature at which Babkin *et al.* made their measurements. This line thus corresponds to theoretical predictions of  $\gamma(\phi)$  using the roughening theory by Nozières after adjustment using all experimental data by Wolf, Gallet *et al.* [2, 7]. Without further adjustment, it also fits the various data successively obtained in Moscow at 1.2 K by Babkin *et al.* (crosses and circles) [3] and around 0.4 K by Andreeva *et al.* (triangles) [4]. This figure calls for further remarks.

No renormalization effects, consequently no temperature variation are expected above the angle  $\phi_{c0}$ . We have averaged various groups of points by Babkin, and the remaining scatter indicates an order of magnitude for the probable error bar in these data, as well as an idea of the difficulty of such experiments. Andreeva's data agree very well with the theoretical curve which, at such large angles, is nothing but the experimental curve described by equation (18).

The width of the critical region ( $0 < \phi < \phi_{c0}$ ) is right. It is only below 0.04 rad that an inflexion occurs in the solid curve because the surface feels the smoothing effect of the lattice. This agreement is of particular importance here, because it strongly supports the numerical factor 3 (or 12) which we described above. As predicted above, it is only below  $\phi_{c0} \approx 0.04$  rad that a deviation from equation (18) occurs, and Andreeva's data at 0.4 K agree both with Babkin's data and with our theory at 1.2 K. An extension of Andreeva's measurements to lower angles should show a temperature variation.

Furthermore, the coupling strength  $t_c$  is directly related to the amplitude of the critical variation, which is also right. At least one of the groups of experimental points comes close to the universal value  $\gamma_R$ . Both the value of  $T_R$  and that of the coupling strength  $t_c$  get further experimental support here. Surprisingly enough, we now consider Babkin's experiment as the best present check of the universal equation (5), although, some time ago, Babkin himself considered his results as contradictory to Nozières' theory. It seems to us that he did not use it

correctly, partly because all our experimental data were not available in 1985. It is also very unfortunate that Babkin did not extend his measurements in the whole temperature range from 1.1 to 1.4 K : his method is presumably very well adapted to a first measurement of  $\gamma(\phi = 0, T > T_R)$ , i.e. a first possible check of the square root cusp of the roughening transition.

Let us summarize this part. Our new analysis brings further support to Nozières' theory of the roughening transition, including to the numerical factors found in the limiting angle below which a tilted surface can feel the effect of the underlying lattice. This means that the crossover towards vicinal or stepped behavior should only occur below  $\phi_c \approx a/12 \xi$ , a smaller angle than previously thought. As we shall see in part 4, this crossover should correspond to a drop in the angular variation of the surface stiffness.

#### 4. Further predictions for $\gamma(\phi)$ and $k(\phi)$ .

It is interesting, although more difficult, to risk predictions in the angular range  $0 < \phi < \phi_c$ , so as to obtain the full angular variation of the surface stiffness even at low temperatures. In this angular range, the surface free energy or tension  $\alpha(\phi)$  can be developed as follows [1]

$$\alpha(\phi) = [\alpha_0 + \beta(\text{tg } \phi/a) + \delta(\text{tg } \phi/a)^3] \cos \phi. \quad (19)$$

Here  $\beta$  is the step free energy and we assume that the interaction energy per unit length is  $\delta/d^2$  between neighbouring steps. With the simplifying assumption of a revolution symmetry around the  $\phi = 0$  axis, such a vicinal surface has a stiffness tensor with two coefficients

$$\gamma_{||} = \alpha + \partial^2 \alpha / \partial \phi^2 = (6 \delta / a^3) \phi. \quad (20)$$

$$\gamma_{\perp} = \alpha + (1/\tan \phi) (\partial \alpha / \partial \phi) = (\beta/a) (1/\phi). \quad (21)$$

The coefficient  $\gamma_{||}$  controls the curvature of the surface in a plane perpendicular to the steps. It is the one mostly considered in section 3, and it vanishes linearly with  $\phi$  (it would vanish exponentially in the absence of interactions and it would tend to a constant for  $1/d$  interactions). On the contrary, the coefficient  $\gamma_{\perp}$  corresponds to the bending of steps and diverges. One experimental evidence that a surface is truly vicinal (below the crossover angle) should thus be the observation of a large anisotropy in stiffness (see Fig. 7).

There are two main physical origins for the interaction between two steps on a vicinal crystal surface. They both lead to a positive energy per unit length, i.e. a repulsion, which is proportional to  $1/d^2$  [1, 15]. The first one is the statistical repulsion which arises from the fact that overhangs are very unlikely, so that steps do not cross. Accordingly to Nozières [1], the corresponding limitation in entropy leads to

$$\delta_s \approx (k_B T)^2 / (\beta d^2). \quad (22)$$

The second one is the elastic repulsion. Each step is surrounded by a certain strain field which gives an elastic contribution to the step energy. We need to estimate this contribution first. It has to be negative, since it corresponds to the elastic relaxation of the lattice around the step under the action of a local force doublet  $f$  [1]. The order of magnitude of  $f$  is typically the surface stress  $\pi_s$ , more precisely  $\pi_s a$ , so that an estimate of the strain field is  $\delta \sigma(r) \approx \pi_s a / r^2$  (it has to decrease quadratically with the distance  $r$ ). An integration from the distance  $\xi$  gives an elastic contribution to the step energy of about  $-\pi_s^2 a^2 / E \xi^2$ , where  $E$  is the Young modulus. This is a small quantity. Indeed, one may assume that the surface stress  $\pi_s$  has the same order of magnitude as the surface stiffness  $\gamma$  or the surface tension  $\alpha$ . For two rather different materials like Silicon ( $E \approx 1.7 \times 10^{12}$  and  $\gamma \approx 10^3$  in cgs units) and hcp helium 4 ( $E \approx 3 \times 10^8$  and  $\gamma \approx 0.3$ ) one finds a ratio  $\gamma/Ea \approx 3\%$  and an elastic contribution which is less than 3% of the step energy  $\beta \approx \gamma a (a/\xi)$  since the correlation length  $\xi$  is larger than  $a$ .

Around neighbouring steps, the strain fields overlap and, the elastic energy being quadratic, a cross term appears which Nozières [1] writes as

$$\delta_{el} \approx 2(1 - \sigma_p^2) f_1 f_2 / (\pi E d^2), \tag{23}$$

where  $f_1$  and  $f_2$  are the surface force doublets on each step and  $\sigma_p \approx 1/3$  is the Poisson ratio. It seems to us that the elastic interaction between two steps, even at the small distance  $\xi$ , should be smaller than the elastic contribution to the energy  $\beta$  of a single step, which is itself smaller than the quantity  $\alpha_0 a \approx \gamma_0 a$ .

The magnitude of the statistical interaction obviously increases with temperature, especially when  $T$  approaches the roughening temperature  $T_R$  where  $\beta$  vanishes. It is interesting to compare  $\delta_s$  to  $\delta_{el}$ . Let us suppose that, as in helium, the step free energy vanishes about linearly from  $\beta_0 \approx \alpha_0 a/10$  at  $T = 0$  to  $\beta_0 = 0$  at  $T_R$ . With the same estimates for the force doublets as above, we find a crossover from an elastic to a statistical repulsion around  $T = 6 \times 10^{-2} T_R$ , a somewhat surprisingly low value which should be corrected as soon as the magnitude of the elastic interaction is better known. We used such qualitative reasonings to draw the predictions of figure 6. An arrow indicates the crossover from rough to vicinal behavior. The step-step interaction  $\delta$  can only be measured from the limiting slope very close to  $\phi = 0$ . In the absence of such a measurement, it would be a little dangerous to take figure 6

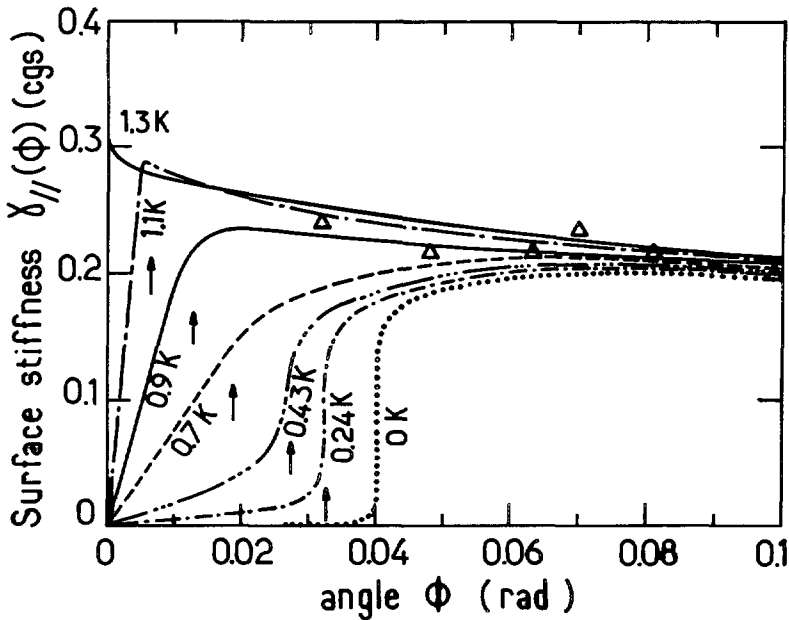


Fig. 6. — A qualitative prediction for the full angular variation of the surface stiffness of crystals near a faceted direction ( $\phi = 0$ ). Below the roughening temperature  $T_R$ , arrows indicate a crossover between a small angle region where steps are well separated (the surface is truly vicinal) to a larger angle region where steps overlap (the surface is truly rough). The crossover angle  $\phi_c$  goes from 0 at  $T_R$  to  $\phi_{c0} \approx 0.04$  rad at zero temperature in the case of helium 4 crystals. The shape of the crossover is not exactly known, and the slope near the origin is mainly due to the statistical repulsion between neighbouring steps. Experimental data obtained around 0.4 K by Andreeva *et al.* [3] have been added for comparison with our predictions. They confirm that no drop of the surface stiffness is observed down to rather small angles (0.03 rad at this temperature), but experiments at lower angle and lower temperature are needed for a more quantitative comparison.

as a precise prediction. It is mostly qualitative also because we only calculated the position of the crossover (there exists no calculation of its shape); furthermore, the extrapolation of Nozières' theory far below  $T_R$  is somewhat dangerous (see Appendix). Still, ignoring the complicated variation of  $\gamma(\phi)$  near  $\phi = 0$  could lead to rather wrong « measurements » of the step-step interactions.

In figure 6, we have added the triangles corresponding to the experimental data obtained around 0.4 K by Andreeva *et al.* [3]. They confirm our prediction that there is no drop of the surface stiffness down to rather small angles (0.03 rad at this temperature), but experiments at lower angles and lower temperatures are needed for a more quantitative comparison.

Let us now briefly consider other possible interactions. At the surface of conducting crystals, there may exist a distribution of electric dipoles on steps. It seems difficult to us to make a precise calculation of the resulting interaction, which is again a repulsion in  $1/d^2$ . In the case of insulating materials such as helium crystals, it can fortunately be ignored.

Helium crystals being in equilibrium with a superfluid liquid, Uwaha recently presented a new type of interaction. Indeed, in this particular case, the fluctuations of steps induce a fluid flow with a certain kinetic energy which contributes to the step free energy. When two or more steps exist, a repulsion occurs from the interference of these flows. According to Uwaha [5], this hydrodynamic repulsion is roughly proportional to  $1/d$  for  $T > 1$  mK and can be written as :

$$\varepsilon_h = I_2 k_B T / (2 \pi^2 d) \quad (24)$$

where the quantity  $I_2$  slowly decays with  $d$  from a value less than one at short distances. A  $1/d$  interaction would lead to a non-zero limit of  $\gamma(\phi)$  when  $\phi$  tends to zero. But Uwaha's new type of interaction is too small to suppress the sharp drop of the surface stiffness which we predict below the critical angle  $\phi_c$ .

Nozières' theory can also be used to calculate how the two coefficients  $\gamma_{||}$  and  $\gamma_{\perp}$  start separating when the angle  $\phi$  decreases below  $\phi_c$ . As explained in reference [1],  $\gamma_{\perp}$  is given by the same expression as  $\gamma_{||}$  except for a change of  $J'_1(u)$  into  $J_1(u)/u$ . We made this numerical calculation whose result is shown in figure 7 for  $T = 1$  K. As expected, the

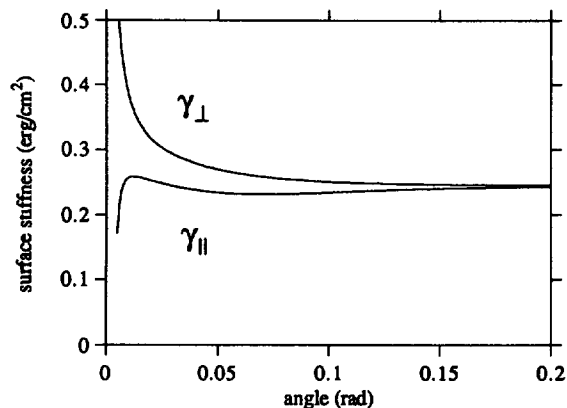


Fig. 7. — A vicinal surface is very anisotropic. Its stiffness tensor has two rather different components.  $\gamma_{||}$  corresponds to a change in the step-step mutual distance, it is the one considered in figures 2, 5 and 6.  $\gamma_{\perp}$  corresponds to the bending of steps. Here, the calculation is made again for helium 4 crystals, the temperature is 1.0 K, a little lower than the roughening temperature  $T_R = 1.30$  K. The two coefficients only separate below the crossover from well separated to overlapping steps, at rather small tilt angle. Their asymptotic behaviour is not drawn and should be governed by the step-step interactions.

surface becomes vicinal, as shown by a strong anisotropy in  $\gamma$ , only below the small angle  $\phi_c \approx 0.01$  rad at this rather high temperature ( $T_R$  is only 1.3 K).

Eventually, one can also calculate the angular variation of the growth rate  $k(\phi)$ , in the limit of zero velocity : then the cut off of the renormalisation depends only on the distance between steps. As again explained in reference [1], one needs to use equation (7) with a modified coefficient. This coefficient  $B^{**}(n, C)$  is given by the same expression as  $A^{**}(n, C)$  except for a change of  $(dx/x)$  into  $dx$ , and for a change of  $2J'_1(u)$  into  $J_0(u)$ . Its value can be read for  $n = 2$  in figure 1, and our final result for  $k(\phi)$  is shown in figure 8. The various curves

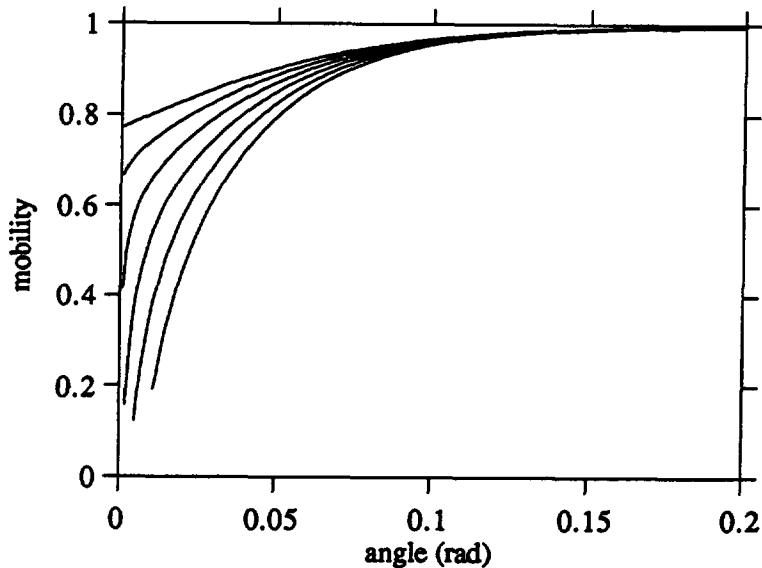


Fig. 8. — A calculation of the angular variation of the growth rate  $k(\phi)$  (or surface mobility) of vicinal surfaces, at the successive temperatures 1.4 K (upper curve), 1.3 K, 1.2 K, 1.1 K, 1.0 K and 0.9 K (lower curve). We have normalized the mobility to that of rough surfaces so that it tends to one at large angle. The departure from the growth rate of rough surfaces takes place at an angle which decreases with temperature and is larger than for the surface stiffness.

correspond to the successive temperatures  $T = 1.4$  K (upper curve), 1.3 K, 1.2 K, 1.1 K, 1 K and 0.9 K (lower curve). Since  $B^{**}$  vanishes at a larger angle than  $A^{**}$ , and because there is no maximum as is the case for  $\gamma_{||}$ , the crossover to vicinal behavior appears at a higher angle, as if the separation of steps affected the growth rate earlier than the surface stiffness. This is in qualitative agreement with the last results of Andreeva *et al.* [3]. However, these authors observed a drop of  $k(\phi)$  at an angle as large as 0.2 rad, a value which is significantly larger than what seems to be predicted by Nozières' theory. They actually also observed that the growth rate of « vicinal » surfaces decreases more rapidly with temperature ( $T^{-5}$  or  $e^{-T/\Delta}$ ) than the growth rate of rough surfaces at larger angle ( $T^{-4}$ ), so that the angular range for an apparent vicinal behavior seems to increase with temperature. Of course their data only correspond to the small temperature range 0.35 to 0.45 K, but, since the width of steps increases with temperature, the vicinal character should be observed inside a smaller and smaller angular window as the temperature increases ( $\phi_c$  decreases). If confirmed, their

observation of an opposite behavior would need some more elaborate interpretations. The study of the angular and temperature variation of the growth rate obviously needs to be extended to lower temperatures and angles.

## 5. Conclusion.

We have shown that for crystal surfaces with a weak coupling to the underlying lattice, a crossover from vicinal to rough behaviour should be observable in the angular variation of the surface stiffness. It corresponds to the change from overlapping degenerate steps to well separated weakly interacting steps. This crossover has not yet been observed but should be observable around a tilt angle  $\phi_c$  which is smaller than previously thought, of order  $a/3w$  where  $a$  is the step height and  $w$  an effective step width. It is only below the critical angle  $\phi_c$  that step-step interactions should be observed and measured.

## Appendix.

In order to keep the reasoning as clear as possible in this article, we omitted the discussion of several difficulties in the use of Nozières' theory. A first major problem is the definition of the step width and its connexion to the height correlation length. This problem is connected with the weak coupling hypothesis which we also need to discuss here.

The profile  $z(x)$ , as defined in equation (1), is a solution of equation (2) with the boundary conditions  $z(-\infty) = 0$  and  $z(+\infty) = a$ . However equation (2) results from a development to leading order in  $\partial z/\partial x$ , a quantity which is small only if the coupling is small, i.e. if the ratio  $V/\gamma$  is small enough. Equivalent conditions are that the step width is large enough or that the step free energy is small enough, and Nozières indeed shows that

$$\xi = (a/2\pi)(\gamma/V)^{1/2} \quad (25)$$

$$\beta = (4a/\pi)(\gamma V)^{1/2} \quad (26)$$

We use equations (25) and (26) for the comparison with experiments. More precisely, and since  $V = UA^2$ , Nozières finds

$$\xi = (1/2 A_{\max})(X_{\max}/Y_{\max})^{1/2} \quad (27)$$

$$\beta = (2 k_B T A_{\max}/\pi)(X_{\max} Y_{\max})^{1/2} \quad (28)$$

where  $A_{\max}$  is the maximum scale at which renormalization is stopped, and  $X_{\max}$  and  $Y_{\max}$  are the maximum values of  $X = 2\gamma a^2/(\pi k_B T)$  and  $Y = 2\pi U/k_B T$ , respectively. In order to get the exact value of  $\xi$  and  $\beta$  from equations (27) and (28), the choice of the renormalization cutoff  $A_{\max}$  seems crucial. This is a delicate point which we want to discuss here, although it might appear to raise criticism about our own work.

Above  $T_R$ , the renormalization does not need to be stopped at any finite scale,  $X$  tends to a finite value and  $Y$ , consequently  $\beta$ , tends to zero. But, below  $T_R$ ,  $Y$  diverges and the theory is no longer valid if it becomes too large, since the coupling becomes too strong. In this article, as in reference [7], we stopped the renormalization when  $U = kT$ , i.e.  $Y_{\max} = 2\pi$ . With this cutoff and close to  $T_R$ , we obtain  $X_{\max} \approx 4$  and

$$\beta \approx 3.2 k_B T A_{\max} \quad (29)$$

$$\xi \approx 0.41/A_{\max} \quad (30)$$

$$\beta \xi \approx 1.3 k_B T. \quad (31)$$

We checked that the value of  $\beta$  is almost independent (within 4 %) of the cutoff in the range  $2 < Y_{\max} < 8$ . This is very useful, since the step energy is a physical quantity which one measures. Unfortunately this is not true for the correlation length  $\xi$  which is not precisely defined, as noticed by Nozières [1]. As a consequence, the step width is not well defined, whether one considers  $\xi$  or  $w = 4 \xi$ , so that the number given in equation (17) should not be considered as an accurate measurement.

The physical interpretation of this cutoff being that the renormalization is stopped when the lattice potential is large enough to kill the surface fluctuations, our choice  $U = k_B T$  looks logical. Since the result is not very sensitive to the exact value of the cutoff, the picture seems rather satisfactory.

However, a delicate problem is raised by Nozières in his lecture notes [1]. By calculating the number of modes and the unit cell area, he obtains that the weak coupling hypothesis does not mean  $U < k_B T$  but  $U < k_B T/4 \pi$ . It thus appears that we are integrating the trajectories up to a region where the weak coupling hypothesis is already violated by one order of magnitude. Our results would not be significantly different if we stopped at  $U = k_B T/3$ , but it appeared impossible to fit them if we stopped the renormalization at  $U = k_B T/4 \pi$ . For example, if adjusted with the step energy, the values of the parameters would not allow a reasonable fit of the growth rate. Although this situation is somewhat puzzling, we thus decided to keep the same cutoff choice as in reference [7], i.e.  $U = k_B T$ .

We want to stress here that the choice of a cutoff is needed only in the low temperature region, in order to compare our measurement of the step energy with Nozières' theory. This is not the case for the measurement of the surface tension and the growth rate.

This discussion is also an opportunity to consider the coupling strength at the microscopic scale. From equations (5) and (11), and from  $t_c = [2 \pi^2 (A(2))^{1/2} U_0] / (\gamma_0 a^2) = 0.58$ , we can infer  $U_0 = 0.058 k_B T_R$ . This means that, close to  $T_R$ , the coupling is indeed weak for  $c$  surfaces of helium crystals, even at the microscopic scale, and even with the most restrictive condition  $U < k_B T/4 \pi$ . This is why, in the integration, we identified the unrenormalized and *microscopic* value  $\gamma_0$  of the surface stiffness with its *macroscopic* value either at high temperatures or away from  $\phi = 0$ . This delicate point might be questionable and was not clearly mentioned in our previous publications. Furthermore, in order to draw the low temperature curves in figure 6, we have extrapolated Nozières' theory down to  $T = 0$  by assuming that one can keep the same values for  $U_0$  and  $\Lambda_0$  in the whole temperature range. This is certainly also questionable and it adds further uncertainties on the exact shape of the low temperature curves on this figure. We still believe that they are qualitatively correct.

Indeed, from the same extrapolation and using equation (26), we can estimate an order of magnitude of the step energy at  $T = 0$ . We find

$$\beta_0/a = 0.07 \quad \gamma_0 = 0.017 \text{ erg/cm}^2 \quad (32)$$

This is the same order of magnitude as the experimental value  $0.02 \text{ erg/cm}^2$  found by Wolf *et al.* [2] from the equilibrium size of  $(10\bar{1}0)$  facets, which shows at least that our extrapolation is not absurd. To find that the step energy is small compared to the surface energy means that the equilibrium size of facets is small compared with the average radius of a crystal ; it is also a more physical evidence that, in helium, the coupling of crystal surfaces to the underlying lattice is small.

Finally, one now knows [1] that, below  $T_R$  and for smooth surfaces ( $\phi = 0$ ), the height-height correlation function saturates around a certain distance, the correlation length, which is  $1/\Lambda_{\max}$  in Nozières' theory. According to equation (30), which is a consequence of the choice  $Y_{\max} = 2 \pi$ , the length  $\xi$  is about half the correlation length  $1/\Lambda_{\max}$  which characterizes the



decay of height fluctuations. A measurement of the height-height correlation function could thus help clarifying the delicate question of the choice of the large scale cutoff in the use of Nozières' theory.

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