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Stepped Surfaces of hcp Helium-4 Crystals

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We have studied melting-freezing waves on helium-4 crystal surfaces which are tilted by a small angle ϕ with respect to the c facets. We present the first experimental evidence that a crossover exists from stepped to rough behavior as a function of angle. We demonstrate this by the independent measurement of the two components $\gamma_\parallel$ and $\gamma_\perp$ of the surface stiffness. In the stepped regime, below the crossover angle $\phi_c \approx 3°$, we also measured both elastic and entropic contributions to the step interactions.

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If tilted by a small angle with respect to a smooth face, a vicinal crystal surface is called "stepped" since it can be described as a set of terraces limited by steps. Such stepped surfaces are expected to be rather anisotropic, with different properties across and along the steps. In particular, the two principal curvatures should be very different on the equilibrium shape of a stepped crystal surface. As realized ten years ago, a crucial quantity in this shape is the nature of the interactions between steps [1]; moreover, step interactions also affect the fluctuations of terrace widths [2], and eventually the roughening transition of vicinal surfaces [3]. Consequently, to calculate and measure step interactions is an important challenge in the physics of crystal surfaces.

The standard description of stepped surfaces supposes that steps are well separated fluctuating lines with weak interactions. However, it was recently pointed out [4] that this description should break down if the step density is too large, i.e., if the tilt angle ϕ is larger than some critical value ϕ_c. Indeed, steps have a certain statistical width and if they are too close to each other (above ϕ_c) they slightly overlap and terraces are not wide enough to be well defined. As a function of angle, a crossover from stepped to rough behavior was thus predicted to occur around ϕ_c [4–6]. It was further noticed [4] that step interaction measurements could only be compared with theory if done at low enough step densities (for $\phi \ll \phi_c$). We hope that the general understanding of this whole problem is improved by our study of vicinal surfaces on helium-4 crystals.

Although most existing theories predict a repulsion proportional to $1/d^2$, where $d = a/\tan\phi$ is the average distance between steps and a their height, no clear experimental evidence had yet been obtained for such an interaction law (for a review, see [7] and references therein). The physical origin for such a $1/d^2$ repulsion can be elastic (the overlap of strain fields around neighboring steps), entropic (steps do not cross each other since overhangs are unlikely), or dipolar (on metallic surfaces). However, disorder could change this law [8] to $1/d$ and other possible mechanisms have also been proposed [9]. Moreover, various experiments led to contradictory results. Some authors [10] tried to measure the equilibrium shape of Pb, In, or He crystals near facet edges. Indeed, a $1/d^2$ repulsion implies a ($z \approx x^{3/2}$) profile equation, although a $1/d$ repulsion would lead to $z \approx x^2$. However, the exact location of the facet edge is very difficult, and it is hard to distinguish between the two possible shapes which were both apparently observed. Wang *et al.* and Alfonso *et al.* [11] later measured the width w of the distribution of terrace sizes on Si surfaces; unfortunately, w depends only weakly on the nature of interactions ($w \approx d$ for $1/d^2$ and $w \approx d^{3/4}$ for $1/d$ interactions) so that a definite conclusion was hard to draw [7]. Finally, the case of Cu $(11n)$ surfaces also seems unclear, since $1/d^2$ repulsions had been observed from the study of roughening transitions [3], but attractive interactions were found from STM studies [12].

With helium-4 crystals, step interactions can be measured from another type of experiment. Indeed, their fast growth dynamics allows the propagation of melting-freezing waves [13], a macroscopic phenomenon from which one can obtain the surface stiffness γ. The surface stiffness is the quantity which controls the equilibrium curvature of crystal surfaces [1]; it is a tensorial quantity related to the surface free energy or surface tension α through the relation $\gamma_{ij} = \alpha + \partial^2\alpha/\partial\phi_i\partial\phi_j$. For stepped surfaces, Nozières [6] called $\gamma_\parallel$ and $\gamma_\perp$ the two principal stiffness components (the tensor is diagonal in the right reference frame). Now the component $\gamma_\parallel$ is proportional to the step interactions, and a $1/d^2$ repulsion should correspond to a linear vanishing of $\gamma_\parallel$ when the tilt angle ϕ tends to zero. Such a measurement was attempted recently by Andreeva, Keshishev, and Osip'yan [14] but it triggered an additional controversy [4,15]. Indeed, Andreeva, Keshishev, and Osip'yan [14] observed a rise in $\gamma_\parallel$ and Andreev concluded that facets do not really exist on helium crystals [15]. Balibar, Guthmann, and Rolley [4] replied that Andreeva's experiments should be extended to lower temperature and smaller tilt angle for a clear evidence of a stepped behavior and a reliable measurement of step interactions to be obtained. This is what is presented here.

We studied the propagation of melting-freezing waves between 40 and 400 mK and for tilt angles ϕ from 0.5° to 6°. We first showed the existence of a crossover from

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stepped to rough behavior around the small angle $\phi_c \approx 3°$. From the asymptotic behavior of the two surface stiffness components $\gamma_{\parallel}$ and $\gamma_{\perp}$ at even smaller angle ($\phi \leq 1°$), we measured the step energy β and the total interaction δ between steps, which is found to be in very good agreement with the $1/d^2$ law. Furthermore, from the temperature variation of this interaction we obtained the respective magnitude of the elastic and entropic interactions $\delta_{\rm el}$ and δ_S which are about equal at 130 mK. We finally compared our results with the recent prediction of a "universal Gaussian curvature" by Akutsu, Akutsu, and Yamamoto [16].

We grow our crystals in a box, inside a cell attached to a dilution refrigerator with optical access through four sets of large windows along the two perpendicular axes *Ox* and *Oy*. Since the box is much larger (24×38 mm^2) than the capillary length (≈ 1 mm), the interface between the crystal and the superfluid above is horizontal, which minimizes gravitational energy. We thus prepare vicinal surfaces with an adjustable tilt by the same method as Andreeva, Keshishev, and Osip'yan. We first nucleate a seed which falls down to the bottom of the box with a "*c*" facet roughly horizontal, i.e., with the sixfold symmetry axis of the hcp structure roughly vertical; we then rotate the box with the crystal inside, in order to orient the lattice with respect to the horizontal surface. This is achieved by two micromotors around two perpendicular axes [17], and the *c* axis is first aligned vertically within 10^{-4} rad; we then rotate the crystals by up to $\pm 6°$ around *Ox* or *Oy*, so that steps are created either perpendicular or parallel to the wave vector *k*. The two surface stiffness tensor components are thus measured independently. The dispersion relation for melting-freezing waves is

$$\omega^2 = [\rho_L/(\rho_L - \rho_c)^2][\gamma k^3 + (\rho_L - \rho_c) g k]\,, \qquad (1)$$

where ρ_L (respectively ρ_c) is the liquid (respectively crystal) density and g the gravity [18]. According to Nozières' notations, the surface stiffness γ takes the value $\gamma_{\parallel}$ if the wave vector k is parallel to the projection of the c axis on the vicinal surface (i.e., perpendicular to the steps) and $\gamma_{\perp}$ if k is perpendicular to c (parallel to the steps). We use frequencies from 0.6 to 3 kHz so that the gravity term in Eq. (1) is only a small correction when calculating γ from ω and k.

Plane waves are excited with the ac electric field of a capacitor evaporated on a glass plate, as done by Wang and Agnolet [19]. The plate is parallel to *Ox* and *k* along *Oy*. The waves are recorded by scanning the surface with a laser beam which is totally reflected at grazing incidence (4° from below). The reflection direction is modulated by the wave and measured with a two element photodiode and a lock-in amplifier. The wave amplitude corresponds to a modulation of the surface orientation which is much smaller than the tilt angle (Fig. 1). The actual height modulation is also very small (here from about 1200 to 200 Å, peak to peak, left to right). In this Letter, we focus on the measurements of the surface stiffness from the wavelength $\lambda = 2\pi/k$. Since k enters Eq. (1) to the third power, an accurate measurement of λ is necessary. This is best done by fitting the wave phase with a cosine function after dividing curves such as in Fig. 1 by the wave amplitude which is also acquired by a computer from the lock-in. The analysis of the damping will be published later; here we only note that it is proportional to the growth resistance which increases when ϕ tends to zero (there are less steps) but fortunately decreases when T tends to zero (the step mobility increases). Measurements below 2° and above 300 mK appeared rather difficult. Also delayed for a later publication is the possible influence of ^{3}He impurities. Here we used ^{4}He with its natural purity ($[^3\text{He}]/[^4\text{He}] = 1.3 \times 10^{-7}$).

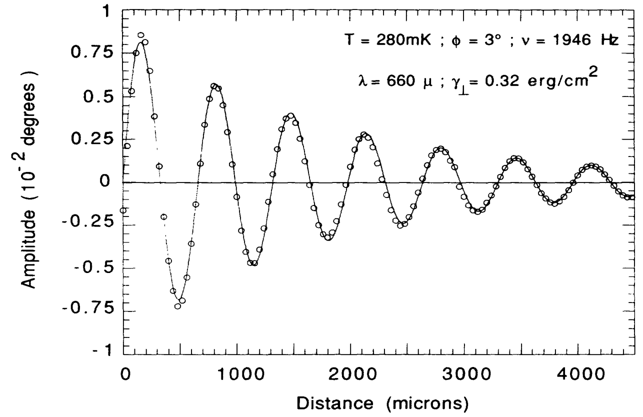

FIG. 1. Circles: typical recording of a melting-freezing wave propagating at the crystal surface. Solid line: best fit with an exponentially damped cosine function.

Results from about 300 recordings corresponding to a dozen of successive crystals are summarized in Fig. 2. As can be seen, the surface properties change as a function of orientation. Above 4°, our low temperature data agree with earlier results obtained by Wolf *et al.* [20] at higher temperature: the surface is rough and nearly isotropic ($\gamma_{\perp} \approx \gamma_{\parallel}$). On the contrary, at smaller angle, a large anisotropy is found: $\gamma_{\perp}$ diverges while $\gamma_{\parallel}$ vanishes at small ϕ. This effect is observed for the first time and it is characteristic of stepped surfaces. In our case, stepped surfaces only exist below the crossover angle $\phi_c \approx 3°$. Balibar, Guthmann, and Rolley [4] explained why such a small angle is expected when the coupling of the crystal surface to the underlying lattice is weak (for metal to vacuum interfaces ϕ_c would be larger). More surprising is the large crossover width, which would be interesting to calculate. As for the results at higher T and ϕ by Andreeva, they show no particular divergence, but only the small anisotropy of the crystal at large angle, which is nonsingular, i.e., not related to steps nor to the c facet.

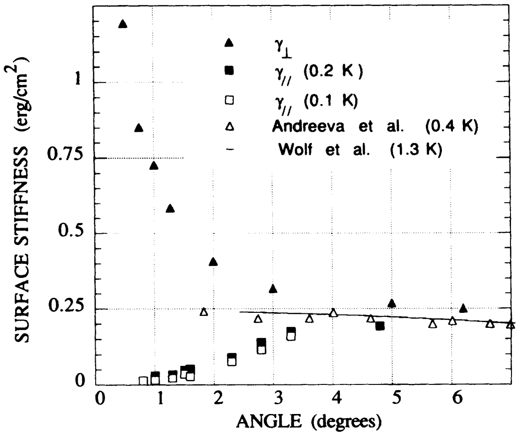

FIG. 2. The two components of the surface stiffness as a function of the tilt angle ϕ. As ϕ tends to zero, $\gamma_\perp$ diverges while $\gamma_\parallel$ vanishes. Above 4°, our low temperature measurements agree with other data obtained at higher temperature by Andreeva, Keshishev, and Osip'yan [14] and Wolf *et al.* [20]: a crossover from stepped to rough behavior has occurred.

Let us now consider the divergence of $\gamma_\perp$. For small ϕ, if steps have a height a and free energy β, if they repel each other with an energy δ/d^2 per unit length, and if α_0 is the surface tension of terraces, one can write [6] the surface tension of a stepped surface as

$$\alpha(\phi) = [\alpha_0 + (\beta/a)\tan\phi + (\delta/a^3)\phi^3]\cos\phi\,. \tag{2}$$

To write Eq. (2), we supposed a rotation symmetry around the c axis, so that only one angular variable remains. The two components of the surface stiffness tensor are thus

$$\gamma_\parallel = \alpha + \partial^2\alpha/\partial\phi^2 = (6\delta/a^3)\phi\,, \tag{3}$$

$$\gamma_\perp = \alpha + (1/\tan\phi)\partial\alpha/\partial\phi = (\beta/a)(1/\phi)\,. \tag{4}$$

As shown in Fig. 3, the stepped behavior is well established below 1° only, where our results are in good agreement with Eqs. (3) and (4). Up to 200 mK, we observed no significant temperature variation of $\gamma_\perp$ and deduced the value $\beta/a = (11 \pm 1.5)\times 10^{-3}$ erg/cm^2 in the $T=0$ limit. As expected in the weak coupling situation [4], this is much smaller than the average surface tension [21] of the crystal ($\beta/a \ll \alpha \approx 0.2$ erg/cm^2). Similarly, one expects the kink energy ε to be much smaller than the step energy per lattice spacing: $\varepsilon \ll \beta a = 10^{-17}$ erg$=$70 mK. Consequently, at the temperature of our experiments, the kink density is very high and the step free energy β is isotropic and identical to the step stiffness. This justifies the above assumption of a rotation symmetry. We also verified that no systematic variation of β could be observed as a function of the step orientation in the c plane.

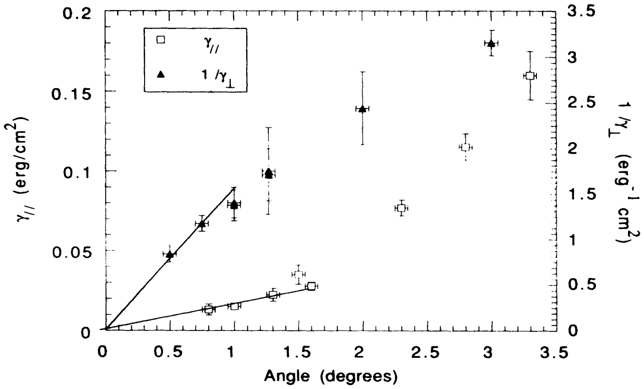

FIG. 3. The measured values of $\gamma_\parallel$ tend linearly to zero below 1.3°, in agreement with $1/d^2$ step interactions. The total interaction δ is measured from the initial slope. Similarly, the step free energy β is obtained from $1/\gamma_\perp$ [see Eqs. (3) and (4)].

Let us finally consider $\gamma_\parallel$. It tends linearly to zero below about 1.3°, in very good agreement with the assumption of $1/d^2$ interactions (with $1/d$ interactions, $\gamma_\parallel$ would tend to a constant at $\phi=0$). Despite difficulties in making measurements at the lowest angles, we measured the step interaction energy δ from the initial slope of $\gamma_\parallel$. According to Akutsu, Akutsu, and Yamamoto [16], the entropic interaction has a temperature dependent magnitude $\delta_S = (\pi k_B T)^2/6\beta$. As for the elastic interaction between two steps, it was written [6] as $\delta_{\rm el} = (2/\pi)f^2 \times (1-\sigma^2)/E$ where $E = 3.05\times 10^8$ cgs is Young's modulus [22], $\sigma = 1/3$ is Poisson's ratio, and f is the amplitude of the force doublet on each step. Since other possible interactions [9] should be much smaller, we fitted the observed temperature variation of δ (Fig. 4) with the interpolation formula [2,23]

$$\delta = (\delta_S/4)[1 + (1 + 2\pi^2\delta_{\rm el}/3\delta_S)^{1/2}]^2\,. \tag{5}$$

We found $\delta_{\rm el}/a^3 = 0.045 \pm 0.007$ erg/cm^2 which gives a value $f/a = 0.85 \pm 0.06$ erg/cm^2. This is close to the value 0.6 estimated by Edwards, Pettersen, and Badder [21] for the surface stress. As for the entropic interaction we found $\delta_S/a^3 = (4.0 \pm 0.7)T^2$ erg/cm^2. Writing it as $A(k_B T)^2/\beta$, we found $A = 1.8 \pm 0.6$, in good agreement

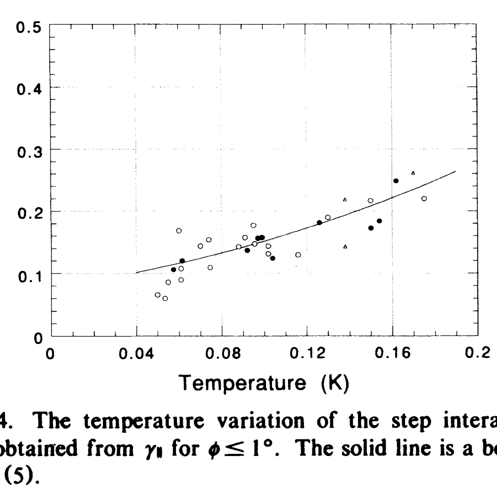

FIG. 4. The temperature variation of the step interaction δ/a^3 as obtained from $\gamma_\parallel$ for $\phi \le 1°$. The solid line is a best fit with Eq. (5).

with Akutsu's value $\pi^2/6 = 1.645$. Of course, the large error bar in A reflects the scatter of data which should be improved (Fig. 4). It is interesting to remark here that the entropic interaction has also been calculated [1,2, 6,23] in terms of the step diffusivity and kink energy, an alternative approach. Translating various equations in Ref. [2] into our notations leads to $A = \pi^2/12$, half what is predicted in Ref. [16] and also found in our experiment. It seems that an unfortunate numerical error in Ref. [23] has led to this apparent contradiction [24].

Finally, Akutsu, Akutsu, and Yamamoto [16] also predicted the existence of a "universal Gaussian curvature" from the relation $(\gamma_\parallel \gamma_\perp)^{1/2} = k_B T \pi / a^2$. It was later noticed [25] that this is only true in the limit where entropic interactions are dominant but that it is violated near $T = 0$ where only elastic interactions remain. We found that the crossover from elastic to entropic interactions occurs around 130 mK. We understand this rather low temperature (the roughening temperature T_{R0} of c facets is 10 times larger) as another consequence of the weak coupling which makes the step energy small and the step fluctuations easy.

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