Devil's Staircase of Facets on the Surface of ^{4}He Crystals

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According to Landau, at $T = 0$ the equilibrium crystal surface consists of an infinite number of facets lying in all directions with rational Miller indices—the so-called devil's staircase phenomenon. We have discovered 11 new types of facets on the surface of ^{4}He crystals, in addition to the three observed before. Some of the new facets are of very high order, lying at angles as small as 4° to the basal c facet, thus forming the predicted devil's staircase. The estimated step energies depend rather weakly on interplanar distance which we explain by the strong anisotropy of the steps.

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Faceting is probably the most exciting phenomenon taking place on the crystal surface. Facets reflect the discrete nature of the crystal, and they are stabilized by the periodic potential induced by the crystal lattice. Ordinary crystals typically show only a few types of facets because the surface phenomena are masked by the bulk effects such as mass and heat diffusion, so that only basic, low order facets can manifest themselves. In contrast, helium crystals at low temperatures present a unique model system to study crystal surfaces, because the latent heat is very small and the liquid phase surrounding the crystals is superfluid, providing fast mass and heat transport.

Landau was the first to recognize that the physical reason for the appearance of a facet on the crystal surface is the discontinuity of the derivative of the surface tension $d\alpha/d\phi$ at the corresponding orientation [1]. Landau considered the surface vicinal to the atomically flat facet as a set of terraces separated by steps. If the energy of the step is positive, the surface tension increases linearly with the absolute value of the tilt angle $\alpha(\phi) = \alpha_0 + \beta|\phi|/d$ (β and d are the step energy and height, correspondingly) and has a cusp at the orientation of the facet. The equilibrium size L of the facet is proportional to the value of the break $d\alpha/d\phi|_{\phi=+0} - d\alpha/d\phi|_{\phi=-0}$ or, to the step energy, $L \sim \beta/(\kappa\alpha d)$, where κ is the curvature of the rough surface close to the facet [1]. A facet of high order consists of terraces of a basal facet separated by equidistant steps. The (secondary) step on such a high order facet appears as a change of the width of a single terrace by one interatomic distance. Landau has shown that the free energy of such a secondary step is finite because of the interaction between primary, basal steps. Thus, at low temperatures the crystal surface in equilibrium consists of an infinite number of facets at any rational direction, that is, the so-called devil's staircase phenomenon [1].

To date, in ^{4}He crystals only three types of facets—$[0001]$ (c), $[10\bar{1}0]$ (a), and $[10\bar{1}1]$ (s)—have been observed long ago by groups in Haifa, Moscow, and Paris using optical cryostats [2]. High order facets have small step energies, and, in equilibrium, they can be observed only on the surfaces which have very small curvature. However, facets with a size much larger than the equilibrium one can be observed on slowly growing crystals. Atomically smooth facets grow by the spiral motion of steps provided by screw dislocations—the so-called spiral growth. At small enough overpressures, the facet is immobile, while the rough surfaces near the facet grow easily and reach the plane of a facet which thus increases in size. Because of the finite size of the crystal, there is always a Laplace overpressure on the facets: $\delta p_F = \alpha\kappa\rho_L/\Delta\rho_{\mathrm{SL}}$. Thus the high order facets with small step energies should be searched on the top of large, very slowly growing crystals.

In this Letter, we report on our investigations on hcp ^{4}He crystals carried out with a low temperature Fabry-Pérot interferometer [3]. An original histogram method has been used for detecting flat areas on the crystal surface with low average curvature. As a result, 11 new types of facets have been observed on the surfaces of slowly growing crystals, and the step energies of the new facets have been estimated.

The interfering light which passes through a crystal gains a phase shift proportional to the crystal thickness. A facet is seen in an interferogram as a set of parallel equidistant fringes, which corresponds to a linear change of the crystal height. In Fig. 1 are shown the c and s facets together with two new, $[10\bar{1}2]$ and $[30\bar{3}4]$ facets. However, a simple inspection of the interferograms to detect plane areas is not sufficient to identify high order facets, because they are stable only near the top of large crystals where the rough surface also has small curvature and thus can be mixed with facets. For instance, the new $[10\bar{1}2]$ facet, which is of low order having the 4th largest interplanar distance (step height) d, has relatively sharp edges and is well visible in Fig. 1, while the new high order $[30\bar{3}4]$ facet is very close to the top of the crystal and has rather smooth edges which makes it difficult to distinguish from the rough surface with small curvature.

In order to avoid such ambiguity, we have developed a novel, histogram technique to identify the flat surfaces on the imaged crystals. In this method we find the direction of

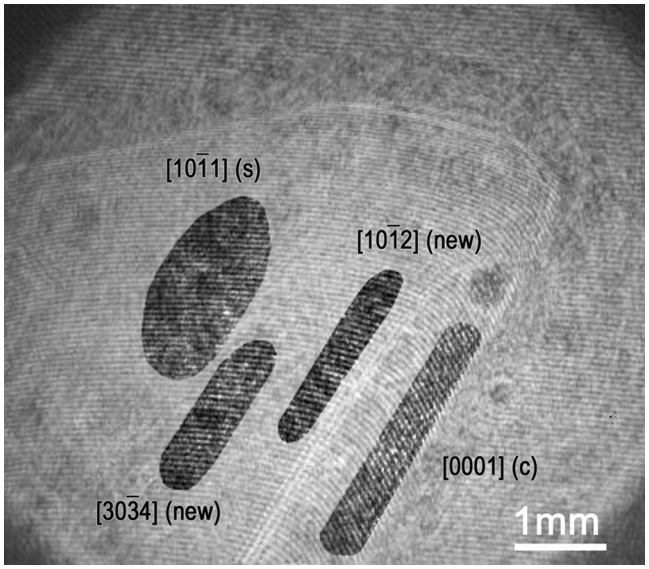

FIG. 1. Interference pattern of a slowly growing ^{4}He crystal at 0.2 K. Marked areas correspond to the known [0001] (c) and [10$\bar{1}$1] (s) facets and to the newly discovered [10$\bar{1}$2] and [30$\bar{3}$4] facets.

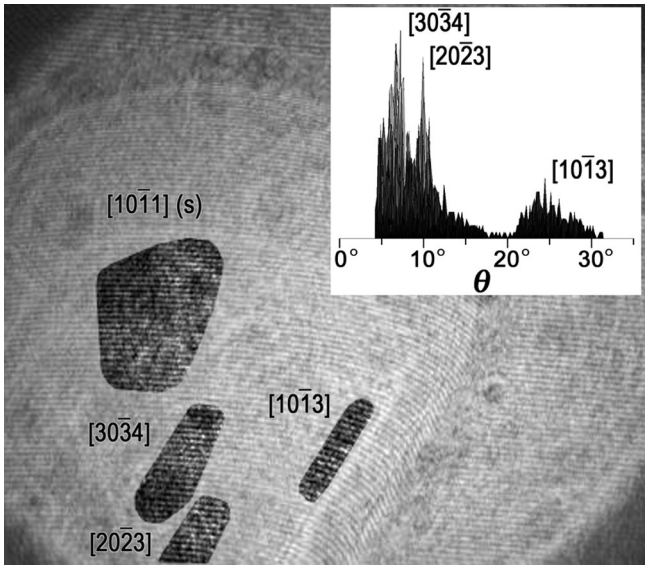

FIG. 2. Original interferogram with marked areas corresponding to the [10$\bar{1}$1] (s) facet and to the newly discovered [10$\bar{1}$3], [20$\bar{2}$3], and [30$\bar{3}$4] facets. Inset: Histogram of the directions of normals to the crystal surface; θ is the angle to the s-facet normal in the plane of s- and c-facet normals.

the normal to the surface at each point of the original interferogram by fitting the phase distribution in the small area around the chosen point with the quadratic form. Then we plot a histogram of the orientations of the crystal surface. In the histogram, a plane facet shows up as a peak at the corresponding direction.

An example of such a histogram, made for the same crystal shown in Fig. 1 but at a different time, is presented in the inset in Fig. 2. The 2D histogram is rotated so that its horizontal axis represents the angle between the surface normal and the normal to the s facet in the plane formed by the normals to the s and c facets. The width of the peak is due to the points of the facet close to its edge, because the fitted areas include there more points which do not belong to the facet and have different orientations. To determine the orientation of the facet more accurately, we have selected the area which contributes to a certain peak and fitted it separately with a linear form representing a plane facet. In this way, we were able to reach the accuracy of about 0.5° in determining the orientation of facets on the crystal surface. A good illustration of the histogram method is the facet [10$\bar{1}$3], which can be easily seen on the interferogram (Fig. 2) and which gives a significant peak in the histogram (the inset in Fig. 2). Two other peaks on the histogram correspond to two high order facets: [20$\bar{2}$3] (10.6° to the s facet) and [30$\bar{3}$4] (7.3° to the s facet).

Note that one and the same facet can be quite large or too small to be seen on the same crystal at different moments during growth of the crystal; for example, the [10$\bar{1}$2] facet is large in Fig. 1 but is not seen at all in Fig. 2, where the [10$\bar{1}$3] facet is present instead. The reason is that the faceted crystal is far from equilibrium even during very slow growth. Growing facets quickly decrease in size, while the size of slowly growing or immobile facets increases. When fast facets become small in size, the slow facets start growing if we continue pressurizing, and in this way different facets appear, disappear, and reappear during growth.

In order to observe very high order facets of type [10$\bar{1}N$], we have created a crystal with an almost horizontal [0001] c facet and grew it very slowly. An example of an original interferogram and the corresponding histogram are shown in Fig. 3. The 2D histogram is rotated so that the horizontal axis on the histogram shows the angle between the surface normal and the normal to the c facet in the plane formed by the normals to the c facet and the lower, [10$\bar{1}$0] a facet. The peaks correspond to very high order facets laying at angles as small as 11.7°, 7.0°, and 4.7° with respect to the c facet. These facets have normals lying on the plane of the [0001] c facet and the lower, [10$\bar{1}$0] a-facet normals, and they can be identified as [10$\bar{1}$9], [10$\bar{1}$15], and [10$\bar{1}$23].

Moreover, the angle as small as 3.5° was obtained for a large facet lying between c and the right, [01$\bar{1}$0] a facet, and the facet has thus been identified as [01$\bar{1}$(32 ± 1)]. This facet produces the largest peak in the histogram, but the corresponding area has been excluded from the histogram shown in the inset in Fig. 3, because this peak is not in plane with other peaks. To our knowledge, facets with such a high Miller index have never been observed before. Facets with Miller indices up to $N = 10$ have been seen in liquid crystals [4] and up to $N = 5$ in bcc ^{3}He crystals [5].

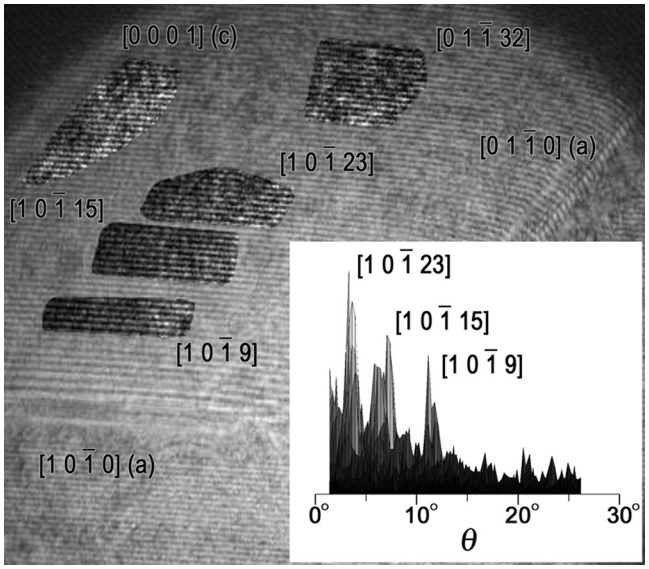

FIG. 3. Original interferogram with marked areas corresponding to the [0001] c facet, [$10\bar{1}0$] a facet, and [$01\bar{1}0$] a facet and to the newly discovered very high order facets (see text). Inset: Histogram of the directions of normals of the crystal surface; θ is the angle to the c-facet normal in the plane of the [0001] c facet and lower [$10\bar{1}0$] a-facet normals.

The observation of such a multitude of facets means that the step energy of facets decreases rather slowly with the decrease of the step height d (increase of Miller indices). A simple estimate of the step energy can be done by measuring the growth threshold:

$$\delta p_{\mathrm{th}} = 2\beta\rho_L/(\Delta\rho_{\mathrm{SL}} d\bar{l}),$$

where $\bar{l}$ is the average distance between dislocations. The overpressure δp_{th} can be found by measuring the hydrostatic pressure difference and the Laplace pressure difference with respect to the rough surface which can be thought to be in equilibrium because of high mobility. Our results are presented in Fig. 4. Indeed, one can see that the data fall onto very slow, nearly linear dependence $\beta \propto d$, with the exception of the two basic c and a facets ($\beta_a \sim \beta_c$ are not shown on the graph).

The observed weak scaling is unexpected and has not been observed before. A first prediction for the step energies of high order facets has been made by Landau, who has shown that the primary steps repel due to the van der Waals interaction, $U_{s-s} \propto 1/x^3$, which leads to the 5th power dependence of the secondary step energy on its height d [1]. Later, Marchenko and Parshin have shown that primary steps induce the field of elastic deformations in the lattice, and this leads to even stronger repulsion: $U_{s-s} \propto 1/x^2$ [6]. Tsepelin *et al.* have shown that the $1/x^2$ interaction between primary steps results in the 4th power dependence of the secondary step energies, and they have indeed observed such scaling in their experiments with bcc ^{3}He crystals [3,7]. Our data suggest a much weaker dependence.

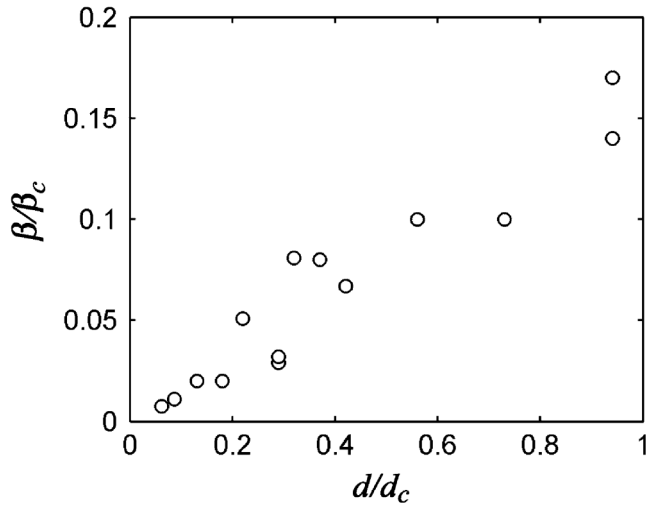

FIG. 4. The measured dependence of the step free energy β on the interplanar distance d for the new facets and for the s facet (two rightmost points).

The observed scaling can be attributed to the anisotropy of an elementary step on vicinal facets. Indeed, the concept of secondary steps discussed above assumes that they are straight, while during spiral growth they are necessarily bent. Let us consider a vicinal facet [$10\bar{1}N$] with a pair of screw dislocations of opposite signs which produce a primary step (Frank-Read source). This additional primary step generates N secondary steps which are pinned to the dislocations (see Fig. 5).

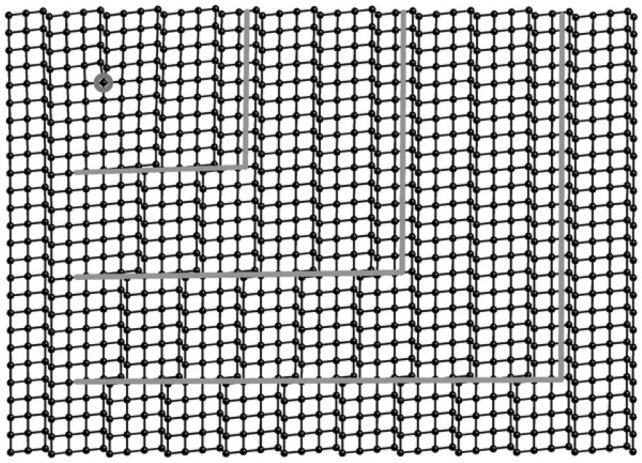

FIG. 5. The high order facet [$10\bar{1}N$] with $N = 3$, normal view. In the top left part is shown a screw dislocation and an additional primary step generated by this dislocation. This step is terminated on the dislocation with opposite sign (not shown) forming a Frank-Read source. The Frank-Read source generates N secondary steps (marked by gray lines) which are pinned to the dislocations until the growth threshold is exceeded. Note the anisotropy of the secondary steps.

As one can see, these secondary steps are highly anisotropic: while the energy of the step oriented parallel to the primary steps $\beta_{||}$ is due to the interaction between them, the energy of the perpendicular secondary step is due to kinks on the primary steps $\beta_\perp = \varepsilon/(Nd_c)$ (ε is the free energy of a kink). If $\beta_\perp \gg \beta_{||}$, the condition for detaching of the secondary step from the dislocations is $d\delta p_{\mathrm{th}}(\Delta\rho_{\mathrm{SL}}/\rho_L)\bar{l} \approx 2\beta_\perp$, similar to the threshold condition for usual spiral growth. In this case, the measured step energies shown in Fig. 4 are actually the energies of the perpendicular steps $\beta_\perp$, which indeed vary linearly with the step height d: $\beta_\perp = \varepsilon d/d_c^2$. From our data we estimate the free energy of the kink: $\varepsilon \approx 0.2\beta_c d_c = 0.2 \times 4.2 \times 10^{-10}$ erg/cm $\times\, 3 \times 10^{-8}$ cm ≈ 20 mK. Note that, because of much weaker scaling, $\beta_\perp \propto d$ will always dominate $\beta_{||} \propto d^4$ at high enough Miller index N. Let us note here that Landau [1] and Tsepelin *et al.* [3,7] have considered only the energy of the parallel secondary steps $\beta_{||}$, which scales much faster with the step height d.

With the increase of temperature, thermal fluctuations suppress the step free energy and set it to zero at a certain roughening transition temperature—the facet disappears. The most developed theory of roughening is the renormalization-group (RG) approach by Nozières and Gallet [8] in which the periodical pinning potential induced by the crystal lattice is renormalized by the surface fluctuations. The RG approach predicts the universal relation between the step height and the roughening temperature

$$T_r = (2/\pi)d^2\sqrt{\gamma_\phi \gamma_\theta}, \tag{1}$$

where $\gamma_\phi = \alpha + \alpha''_{\phi\phi}$ and $\gamma_\theta = \alpha + \alpha''_{\theta\theta}$ are the principal components of the surface stiffness tensor. In our experiments with hcp ^{4}He crystals, we have observed 14 different types of facets in the temperature range of 0.13–0.2 K, while according to Eq. (1) and the measured values of the surface stiffness [9], only 6 types of facets should be present. Thus the current theory fails to explain our findings.

However, according to Fisher and Weeks [10], T_r defined by Eq. (1) is only the lower limit of the roughening temperature, and some other effect could stabilize a facet at higher temperature. Note in this connection that Nozières and Gallet have taken into account only the modulation of the pinning potential in the direction perpendicular to the surface, while in the case of vicinal surfaces there is also a strong modulation of the potential *along* the surface. The concept of secondary steps, in turn, takes somehow into account longitudinal modulation but neglects the surface fluctuations. Thus our findings call for a new theory of roughening which would be able to describe the vicinal surfaces.

In summary, we have observed 11 new types of facets on the surface of hcp ^{4}He crystals, in addition to only three types of facets observed earlier. We have also estimated the step energies of all observed facets, while only the step energy of the basal [0001] c facet was known before. In contrast to theoretical expectations, the dependence of the step energy on the interplanar distance is rather slow, which we explain by the strong anisotropy of the step energy of vicinal facets.

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