On the Growth Dynamics of ^{4}He Crystals near the First Roughening Transition.

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Abstract. The first roughening transition of a surface of hcp ^{4}He crystals was carefully studied in Paris in 1980s. By investigating the growth dynamics of the (0001) facet, the free energy of an elementary step was measured in the close vicinity of the transition and a good agreement was found with the theory of critical fluctuations developed by Nozières and Gallet. We believe, however, that the interpretation of the growth data near the roughening transition made by the Paris group is not self-consistent. We argue that with the step energies they obtained, assuming that the growth is due to the process of 2D-nucleation of terraces, another growth mechanism provided by screw dislocations should be much more effective.

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Facets exist on a crystal surface at low temperatures as smooth planes which are stabilized by the periodic lattice potential. As temperature increases, thermal fluctuations tend to destroy the facets, until the surface enters the rough state at the so-called roughening transition temperature T_R. The order parameter, which determines the size of the facet, is the free energy β of the elementary step on the facet.

However, measurements of the equilibrium facet size are very difficult. Another way to investigate the free energy of the elementary step is to study the growth kinetics of the facet. The growth rate of the facet strongly depends on the step free energy which can be thus measured provided that the growth mechanism and the corresponding kinetic coefficients are known.

Here we will discuss experiments and results reported in two papers, by Wolf *et al.* [1] and by Gallet *et al.* [2] In both papers similar techniques were used to investigate the growth dynamics of the (0001) facet of a hcp ^{4}He crystal near its roughening transition at $T_R = 1.3$ K. The chemical potential difference between liquid and solid phases $\Delta\mu$ was obtained by measuring the height difference H between the sample crystal and the reference crystal with much larger horizontal surface: $\Delta\mu = (\rho_s - \rho_l)gH/\rho_s$. Above T_R the velocity of the (0001) orientation is proportional to the driving force, $u \propto \Delta\mu$, which is the property of the rough surface. Below T_R the dependence of u on $\Delta\mu$ becomes nonlinear, indicating facet formation.

Wolf *et al.* [1] and Gallet *et al.* [2] suggested that the dominant growth mechanism of the facet is spontaneous nucleation of seeds of a new layer, and the growth velocity of the facet varies as $\Delta\mu \exp\left[-\pi\beta^2/(3d\rho_s\Delta\mu k_B T)\right]$, where d is the step height. The values of $\ln(u/\Delta\mu)$ plotted against $1/\Delta\mu$ fall indeed onto a straight line, and from the slope of the line the step free energy was obtained at different temperatures down to 1.130 K.

There is another mechanism of growth which is provided by the steps originating at screw dislocations. If driving force is applied, these steps form spirals which rotate around the dislocations, and the velocity of the facet varies as square of the driving force:

$$u_{sp} = \frac{N\rho_s d^2 k_{st} \Delta\mu^2}{19\beta}. \tag{1}$$

Here k_{st} is the mobility of the step and N is the number of layers produced by one turn around the dislocation. For such a growth there is a certain threshold because the step typically connects two dislocations of opposite sign, forming the so-called Frank-Read source, and cannot move away until the chemical potential difference compensates the largest possible curvature of the step, $2/l$ (l is the size of the Frank-Read source). The corresponding threshold is found from the average distance between dislocations $\langle l\rangle$: $\Delta\mu_c = 2\beta/(d\rho_s\langle l\rangle)$.

Wolf *et al.* and Gallet *et al.* evaluated the velocity of the spiral growth by using Eq. (1). They estimated that under the conditions of their experiments the spiral growth mechanism provides smaller facet velocities than they measured and is thus not relevant. In their evaluation they assumed that the mobility of the step is equal to the mobility of the rough surface. However, the assumption that the mobility of the step is the same as the mobility of the rough surface seems not correct. Only if the step is very sharp, so that the interface changes its height by d over the distance of a few interatomic spacings, then the step mobility can indeed be taken to be approximately equal to the mobility of the rough interface, disregarding

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the peculiarities of the scattering of quasiparticles by the microscopic step. Near the transition, where the step width ξ diverges, the step can be thought as the rough interface tilted by a very small angle $\sim d/\xi$ with respect to the facet orientation [3]. When the chemical potential difference is applied, the velocity of the step *along* the facet is larger by a factor of $\sim \xi/d$ than the *normal* velocity of the rough surface.

In the theory of critical fluctuations developed by Nozières and Gallet [3], the step width ξ and the step mobility k_{st} are given by

$$\xi = \frac{2}{\pi^2}\frac{\gamma d^2}{\beta}, \quad k_{st} = \frac{\pi^2}{2}\frac{\xi}{d}k_R = \frac{\gamma d}{\beta}k_R, \qquad (2)$$

where $\gamma \approx 0.24\,\mathrm{erg/cm^2}$ [1] is the surface stiffness. We can estimate that even at 1.130 K, the lowest temperature of the measurements, the reported value of the step free energy $\beta(1.130\,\mathrm{K}) = 1.4\cdot 10^{-11}$ erg/cm suggests the step width $\xi = 4\cdot 10^{-6}$ cm and the factor of 600 in the mobility of the step compared to the mobility of the rough interface. This factor becomes even larger when approaching the roughening transition, but it was not taken into account by Wolf *et al.* [1] and Gallet *et al.* [2] in their estimation of the velocity of the spiral growth. It is thus clear that the possible role of the spiral growth in these experiments should be revised, as it was already pointed out in the recent review on helium crystal surfaces [4].

To obtain the velocity of the facet growing due to screw dislocations, we merge the mobility of the step given by Eq. (2) into the basic relation (1) and find $u_{sp} = N\rho_s d^3\gamma k_R \Delta\mu^2/(19\beta^2)$. We will take the value of $N = 2$ as the lowest possible for (0001) orientation and use the same value of the rough surface mobility, $k_R = 3.1\cdot 10^{-6}e^{7.8\mathrm{K}/T}$ s/cm, as was used by Wolf *et al.* and Gallet *et al.* The values of the step free energies are taken as they were reported by these authors.

We have found that, at all temperatures and at all driving forces, the velocity of the facet growing by screw dislocations is much higher than the measured velocities. Figure 1 presents three sets of original raw data obtained at 1.130 K, 1.173 K (Wolf *et al.* [1]) and 1.205 K (Gallet *et al.* [2]) in $\sqrt{u}$–$\Delta\mu$ coordinates. The velocities of the spiral growth with the values of the step energy reported in these works, namely $\beta(1.130\,\mathrm{K}) = 1.4\cdot 10^{-11}$ erg/cm, $\beta(1.173\,\mathrm{K}) = 6.3\cdot 10^{-12}$ erg/cm, and $\beta(1.205\,\mathrm{K}) = 8.4\cdot 10^{-13}$ erg/cm, are shown by straight lines which are connected by arrows with the corresponding experimental data sets. One can see that these lines lie always higher than the experimental points, and in some cases the spiral growth with the reported step energies gives an order of magnitude faster growth than it was actually measured.

We can also evaluate the values of the threshold for spiral growth using the average distance between dislocations measured in the same work by Wolf *et al.* [1],

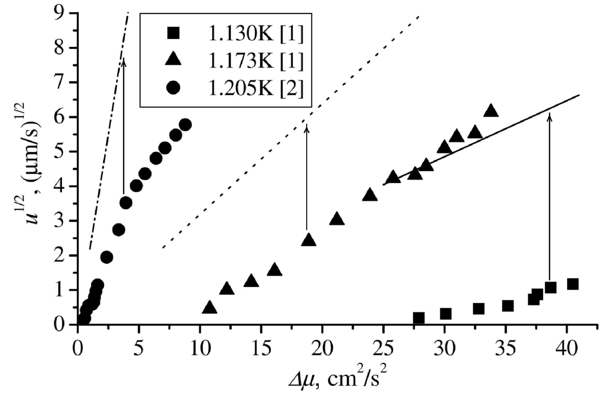

FIGURE 1. Velocities of the (0001) facet measured by Wolf *et al.* [1] and by Gallet *et al.* [2] (symbols) and evaluated velocities of the spiral growth (lines). See text for details.

$\langle l\rangle \approx 0.01$ cm. We find $\Delta\mu_c = 0.5\,\mathrm{cm^2/s^2}$ at 1.130 K, $0.2\,\mathrm{cm^2/s^2}$ at 1.173 K, and $0.03\,\mathrm{cm^2/s^2}$ at 1.205 K, so that $\Delta\mu_c$ is smaller by two orders of magnitude than the actual driving forces applied in the experiment (see Fig. 1).

We conclude that the values of the step free energies found in the experiments by Wolf *et al.* and by Gallet *et al.* suggest much larger facet velocities than what was measured, and hence the results reported in these two papers are not self-consistent. Thus, 20 years after these pioneering works, the adequate understanding of the nature of the roughening transition is still absent, and we would like to renew the attention to this interesting phenomenon.

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