Surface of a ³He Crystal: Crossover from Quantum to Classical Behavior

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³He crystals start to show facets on their surface only at about 100 mK, well below the roughening transition temperature. To understand the reason for that, we have performed the first quantitative investigation on the growth dynamics of the basic (110) facet at 60–110 mK. The obtained values of the step free energy suggest an extremely weak coupling of the solid-liquid interface to the crystal lattice which we show to be the result of quantum fluctuations of the interface. The renormalization group approach by Nozières and Gallet, modified to incorporate quantum fluctuations, explains well the temperature dependence of the step energy measured in this work and at ultralow temperatures by Tsepelin *et al.*, where the coupling is known to be strong. We have thus shown that, paradoxically, the role of quantum fluctuations is at higher temperatures much larger than at low temperature.

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The crystal surface can be either in a rough (rounded) or in a smooth (faceted) state. Facets reflect the periodic nature of the underlying lattice. The measure of the strength of the coupling between the facet and the lattice is the energy of an elementary step which separates the terraces of neighboring atomic layers. At low temperatures the step energy is finite, which causes a strong anisotropy of the surface stiffness and of the growth rate. As a result, a macroscopic facet is present on the crystal surface in equilibrium or during growth. However, at a certain temperature the crystal surface enters a rough state [1], where the step energy is zero and the crystal surface is rounded without any peculiarity. Fisher and Weeks [2] and Jayaprakash *et al.* [3] have found the universal relation between the roughening transition temperature T_R, surface stiffness γ, and step height d: $T_R = (2/\pi)\gamma d^2$.

In bcc solid ³He there exists a big discrepancy between the calculated roughening temperature of the (110) facet, $T_R = 260$ mK, and the highest temperature at which this type of facets has been observed in the experiment, $T_{\mathrm{obs}} \approx 100$ mK [4]. Rolley *et al.* [4] have explained this contradiction by dynamic roughening which blurs the growth anisotropy if an excessive driving overpressure is applied [5]. Domination of dynamic effects indicates that the coupling of the crystal surface to the lattice is weak; Rolley *et al.* have estimated the step free energy β of the (110) facets to be smaller than 10^{-11} erg/cm at 100 mK.

On the other hand, at 0.55 mK the measured step energy of the (110) facets, $\beta_0 = 6.6 \times 10^{-10}$ erg/cm [6], is close to the value of γd, which means that the coupling is strong, as in usual crystals. To understand the nature of this dramatic change (occurring well below the roughening transition temperature), measurements of the step free energy in a wide temperature range were necessary.

In this Letter, we describe our interferometric measurements on the growth dynamics of the (110) facet on bcc ³He crystals. An original method has been used to detect the extremely small (a few Pa) overpressures on the facet by measuring the curvature and the velocity of the adjacent rough parts of the crystal surface. By this way, the free energy of an elementary step was measured in the temperature range of 60–110 mK.

As long as the step energy is responsible for the anisotropy of crystal growth, it can be obtained by measurements on the growth dynamics of a crystal. In the case of ³He far away from the melting curve minimum ($T_{\mathrm{MCM}} = 320$ mK), the mobility of the interface is limited by the transport of the latent heat L, which results in the temperature difference ΔT_{ls} across the moving interface [7]. This difference is compensated by an excess liquid pressure over the equilibrium melting pressure $\delta p = (\rho_s \rho_l / \Delta\rho) L \Delta T_{ls}/T$ [8], where ρ_s and ρ_l are the mass densities of the solid and liquid, and $\Delta\rho = \rho_s - \rho_l$. For a rough surface which grows uniformly, the temperature difference $\Delta T_{ls,R}$ is due to the Kapitza resistance R_K, and the overpressure is proportional to the normal velocity of the interface: $\delta p_R = L^2 \rho_s^2 \rho_l R_K v_R/(\Delta\rho T) \equiv \lambda v_R$ (all the latent heat is released in the liquid) [7].

A smooth facet lacks sticking sites for atoms; therefore in the regime of small overpressures facets can grow due to only the motion of the elementary steps provided by screw dislocations. When overpressure δp_F is applied, the steps move, forming spirals which rotate around the dislocations. The step mobility is also restricted by thermal effects, but, in contrast to the uniform heat flux through the rough surface, the heat flux through the facet has peaks near the moving steps where the latent heat is released. At temperatures near 100 mK the heat conductivity of the solid ³He is by 2 orders of magnitude higher than that of the liquid, and we may consider the temperature of the solid as a constant. By solving the thermal diffusion equation for the set of moving steps separated by the distance l, we find the temperature difference $\Delta T_{ls,\mathrm{st}}$ across the step and the corresponding overpressure on the facet δp_F as functions of the velocity of the steps v_{st}: $\Delta T_{ls,\mathrm{st}} = [R_K/l + \ln(l/w)/(\pi\kappa)]d\rho_s L v_{\mathrm{st}},$

$$\delta p_F = \frac{L^2 \rho_s^2 \rho_l R_K}{\Delta \rho T} v_F + \frac{L^2 \rho_s^2 \rho_l d}{\pi \Delta \rho \kappa T} \ln \frac{l}{w} v_{\text{st}}. \qquad (1)$$

Here κ is the heat conductivity of the liquid, $w \approx \gamma d^2 / \beta$ is the step width [5], and $v_F = (d/l)v_{\text{st}}$ is the facet normal velocity.

The first term on the right-hand side of Eq. (1) has exactly the same form as the overpressure on the rough surface and corresponds to the average heat flux through the interface. The second term corresponds to the heat spread in the liquid near the moving step and thus equals to the effective overpressure δp_{st} which drives the motion of a single step on a facet. By applying the solution of the equation for the spiral growth [Ref. [1], Eq. (42)], we find the relation between the facet velocity v_F and $\delta p_{\text{st}} \equiv \delta p_F - \lambda v_F$:

$$v_F = \frac{\pi \Delta \rho^2 \kappa dT}{20 \beta \rho_s^2 \rho_l^2 L^2 \ln(l/w)} \delta p_{\text{st}}^2, \qquad (2)$$

where $l \approx 20 \beta \rho_l / (d \Delta \rho \delta p_{\text{st}})$ is the asymptotic distance between the neighboring steps of a spiral [1]. The only unknown parameter in Eq. (2) is the step energy β, which can thus be obtained by measuring the velocity-overpressure dependence of a facet.

It is a very difficult experimental task to measure an overpressure directly since it depends on the temperature at the interface, which can hardly be determined with the needed accuracy. Because of the large latent heat and poor heat conductivity of the liquid, significant thermal gradients are always present during crystal growth. Fortunately, the crystal surface itself can serve as a very sensitive, local overpressure gauge. Considering the Laplace pressure of the crystal surface and the variation of the hydrostatic pressure in the liquid, one can express the overpressure in terms of the height h and the curvature K of the crystal surface: $\delta p = \text{const} - \rho_l g h - \rho_l \gamma K / \Delta \rho$.

For a successful experiment, in order to avoid the nucleation of new crystals, the growth velocity of a sample crystal should be smaller than $\sim 0.1 \ \mu\text{m/s}$ [9] and the temperature should not change faster than a few mK/day. Using a cryogenic valve in the ^{4}He filling line of our Pomeranchuk-type cell and a feedback program to stabilize the temperature, we were able to grow a single crystal of about 4 mm diameter and measure its growth dynamics in the temperature range of 60–120 mK. The whole experiment, however, took about 3 months to complete.

To image the crystal surface, we used a high-resolution low temperature Fabry-Pérot interferometer [10]. By fitting the phase distributions over the original interferograms with a polynomial (see Fig. 1), we obtained smooth surface profiles $h(x, y)$ from which the curvatures and velocities of the crystal surface were found. We were thus able to measure not only the growth rate but also the variation of the driving overpressure on the crystal surface by the interferometer only; no other device, such
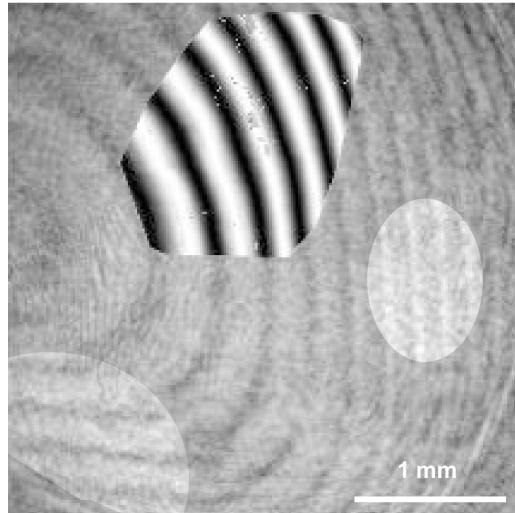

FIG. 1. Interference pattern of a growing ^{3}He crystal at 78.4 mK. The inset is a fit of the phase distribution over the area corresponding to the rough (rounded) surface used as a reference for the overpressure measurements; two (110) facets are seen as sets of parallel equidistant fringes (highlighted) below and on the right-hand side of the inset. The height difference between the surface areas mapped by neighboring fringes is 0.20 mm. The orientation of the selected rough areas is such that they cannot contain (100) or (211) facets which also could be present at these temperatures.

as a thermometer or a usual mechanical pressure gauge, and no calibrations were needed. The accuracy of our method is a fraction of Pa, most of the error being due to the curvature measurements. The effective overpressure on the step is given by $\delta p_{\text{st}} = \lambda(v_R - v_F) + \rho_l g(h_R - h_F) + \rho_l \gamma K_R / \Delta \rho$, where index F relates to facet and R to some (reference) rough part of the crystal surface. The velocity-overpressure dependencies measured at different temperatures are shown in Fig. 2.

The step energies were found by fitting the experimental data with the spiral growth model and are presented in Fig. 3 together with the value measured at 0.55 mK by Tsepelin et al. [6]. We tried to explain the observed temperature dependence of the step energy following the renormalization group (RG) approach developed in detail by Nozières and Gallet [5]. In the RG theory, the step energy is set by the surface stiffness γ and the periodic pinning potential $V \cos(2\pi z/d)$ which represents the interface-lattice coupling. Generally, both γ and V are renormalized by the short-wave thermal fluctuations, but in the first order of the expansion in powers of V/γ the renormalization of γ is negligible, and one may write

$$\beta = (4d/\pi)\sqrt{\gamma V}, \qquad V = V_0 \exp(-2\pi^2 \langle z^2 \rangle / d^2), \quad (3)$$

where V_0 is the bare coupling energy at microscopic scale. The average amplitude of thermal fluctuations of the interface is given by
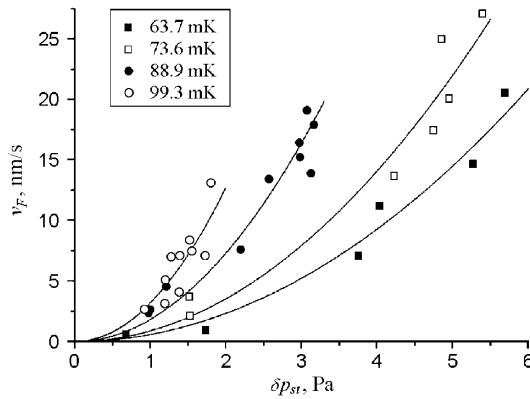
FIG. 2. Velocity of the (110) facet of a ^{3}He crystal at different temperatures as a function of the effective overpressure on the step (see text). Solid curves are quadratic fits to the data.

$$\langle z^2 \rangle = \frac{T}{2\pi\gamma} \ln\frac{k_0}{k_*}, \qquad (4)$$

where k_0 is the short scale cutoff, and k_* is the wave vector corresponding to the scale at which the coupling energy per unit cell, $4\pi V/k^2$, reaches the energy of fluctuations with corresponding k and thus locks the interface at larger scales [5]. As temperature approaches $T_R = (2/\pi)\gamma d^2$, k_* and therefore the step energy drop to zero. We have fitted our data by Eqs. (3) and (4) with the values of V_0 and k_0 being the fitting parameters. The fit is rather good (dashed line in Fig. 3), but the values of fitting parameters are confusing. The bare coupling energy V_0 is by 3 orders of magnitude smaller than the surface tension, giving the zero-temperature value of the step energy 20 times smaller than the value measured by Tsepelin *et al.* [6]. In turn, the value of the short scale cutoff, $k_0 = 10/d$, is certainly too high.

We conclude that the standard renormalization theory [5] fails to explain the experimental data. In fact, this is not surprising since this theory ignores quantum fluctua-
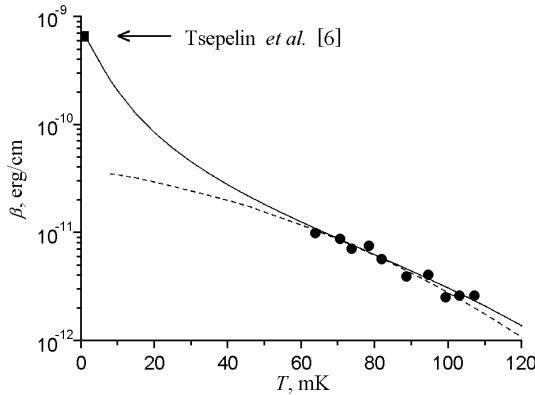

FIG. 3. Measured step free energy of the (110) facet of a ^{3}He crystal as a function of temperature. Dashed line is a fit by the theory of renormalization by thermal fluctuations [5] [Eqs. (3) and (4)]. Solid curve is a fit by the generalized renormalization theory [Eqs. (3) and (6)].

tions, whereas the amplitude of zero-point oscillations of helium atoms is comparable with the interatomic distance and, consequently, the amplitude of quantum fluctuations of the interface could be quite high. Thus, the Nozières-Gallet theory should be modified to incorporate quantum fluctuations.

To do it, we used the dynamic version of the RG theory [5], based on renormalization of the Langevin-type equation of the interface motion

$$\eta\dot{z} - \gamma\nabla^2 z + \frac{2\pi V}{d} \sin\left(\frac{2\pi z}{d}\right) = R(t, \vec{r}), \qquad (5)$$

where η is the intrinsic damping coefficient of the interface and R is the random force. The "inertial" term with second time derivative is negligible due to strong damping. The general form of the correlation function of R is set by the fluctuation-dissipation theorem [11], $R^2_{k,\omega} = \eta\hbar\omega \coth(\hbar\omega/2T)$. We point out that the surface displacement z is calculated from the crystal lattice; thus it is entirely due to the transfer of particles from one bulk phase into another and has nothing to do with the thermal and zero-point oscillations of atoms in the solid.

The renormalization procedure does not differ from the usual one. One starts with the short scale k_0, where the nonlinear "pinning" term in Eq. (5) is assumed to be small and the crystal surface is freely fluctuating. Again, as in the classical case, in the first order only V is renormalized, and we recover Eq. (3). $\langle z^2 \rangle$, after integration over k, can be expressed now as

$$\langle z^2 \rangle = \frac{\hbar}{4\pi^2\gamma} \int_0^{\omega_0} d\omega \coth\frac{\hbar\omega}{2T} \left(\arctan\frac{\gamma k_0^2}{\omega\eta} - \arctan\frac{\gamma k_*^2}{\omega\eta}\right), \qquad (6)$$

where ω_0 is the high frequency cutoff.

Note that in the classical limit the damping coefficient η disappears from the final formula. On the contrary, in the opposite limit of high frequencies, both the value and temperature dependence of $\langle z^2 \rangle$ are essentially determined by the damping. Note also that in the derivation of Eq. (6) we have neglected possible dispersion of η at high frequencies and wave vectors. To our knowledge, this problem was never discussed earlier.

As for the temperature dependence of the damping coefficient, it was discussed by Puech *et al.* [12], who suggested that η varies as $1/T$ due to the Fermi degeneracy of the liquid phase in contact with nondegenerate solid: the "bottle neck" is the energy transfer across the interface. Since this conclusion is based on purely statistical arguments, it seems to remain valid even at very high frequencies, up to the Fermi frequency. In the absence of more elaborate theory, we assumed in our analysis that $\eta \propto 1/T$. As a reference, we have used the value of the damping coefficient measured at the melting curve minimum $\eta(320 \text{ mK}) = 66 \text{ g}/(\text{cm}^2 \text{ s})$ [7]. When fitting the experimental data, we chose the value of the bare amplitude of the pinning potential V_0, which gives the

right value for the step energy at ultralow temperatures, so that we again had only two fitting parameters, ω_0 and k_0, for our set of data.

The generalized renormalization theory [Eqs. (3) and (6)] fits the experimental data very well (solid line in Fig. 3), and the obtained values of the fitting parameters $k_0 = 1.9/d$ and $\omega_0 = 7.4 \times 10^{11}$ 1/s are convincing. Indeed, the value of k_0 is very close to the highest possible wave vector of the oscillations of the [110] plane, $\sqrt{2/3}\pi/d$. The value of ω_0, in turn, agrees well with the frequency range of fluctuations in liquid ^{3}He obtained in the experiments on inelastic neutron scattering [13]. In fact, at 0.55 mK the coupling is strong and the developed approach is valid only qualitatively [5]; hence the obtained values of k_0 and ω_0 probably need some small correction. Note that in the close vicinity of the roughening transition the temperature dependence of the step free energy is determined by the large-scale thermal fluctuations, as in the standard RG approach, while the quantum fluctuations only renormalize the bare step energy to a small but essentially finite value.

Significant reduction (up to 90%) of the bare step energy β_0 due to quantum fluctuations apparently takes place also on the solid-superfluid interface of ^{4}He. However, in that case the effect is not as strong (basically owing to much larger value of the surface stiffness) as in ^{3}He and it is temperature independent [14] because the surface fluctuations (crystallization waves [15]) are weakly damped and their spectrum does not depend on temperature. In contrast, in ^{3}He the damping is relatively strong and quickly increases when temperature decreases, providing the increase of the step free energy. The damping remains strong also below the superfluid transition in the liquid and the antiferromagnetic transition in the solid at 2.5 and 0.9 mK, respectively, at all frequencies except for frequencies below $\sim 10^8$ 1/s, where the superfluid and antiferromagnetic fluctuations take place. Corresponding contribution to the total amplitude of the surface fluctuations can be shown to be small, and thus there should not be significant effect of these transitions on the step energy. On the other hand, a possible ferromagnetic ordering of the interface at $T = 20$–30 mK (see, for instance, Ref. [16]) could increase significantly the value of V_0 at low temperatures.

With the measured values of the step free energy we can find the conditions for dynamic roughening, which takes place when the overpressure on the facet, $\delta p_F \gtrsim \rho_l \gamma K_R / \Delta\rho$, exceeds the threshold for spontaneous nucleation of the seeds of a new solid layer, $\delta p_c \approx \beta^2 \rho_l / (d^3 \gamma \Delta\rho)$ [5]. Thus, the minimum step energy which can be measured with a crystal of a size D is $\beta_{\min} \approx \gamma\sqrt{4d^3/D}$, or about 10^{-12} erg/cm for a crystal with the diameter of 4 mm, and above the corresponding temperature of about 130 mK the crystal shape is always rounded.

To conclude, our measurements and analysis show that in ^{3}He strong quantum fluctuations of the solid-liquid interface keep the step free energy too small to be measured down to temperatures of about $0.4T_R$. To our knowledge, this is an exceptional case. However, when temperature decreases, the quantum motion of the interface with respect to the crystal lattice appears to be more and more suppressed, and the facets show up. Finally, at ultralow temperatures, the surface of a ^{3}He crystal becomes an even more classical object than in ^{4}He—contrary to usual expectations based on comparison of the amplitudes of zero-point oscillations in the bulk solids.

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