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Facet Growth of ^{4}He Crystal Induced by Acoustic Waves

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Very fast growth of the c-facet of a ^{4}He crystal was induced by acoustic waves. The growth velocity was larger at lower temperatures and saturated below about 400 mK. The velocity was proportional to the acoustic wave power. This fast growth cannot be explained by the spiral growth mechanism for the known value of the step mobility. We developed a step multiplication model for high-power acoustic waves and found reasonable agreement with the observed temperature and power dependence of the growth velocity.

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A fundamental problem in crystal growth physics is how fast a facet can grow. In ordinary materials, the facet grows with a spiral growth mechanism well below the roughening transition temperature.[1)] Step motion is limited by the diffusion of atoms and/or by the transport of latent heat. When two steps with opposite sign collide, they disappear without any reflection or transmission. Step density cannot increase indefinitely but reaches a steady-state value which is determined by the driving force for the crystallization. Recently, Parshin and Tsymbalenko have proposed a new type of collision of steps in the case of high mobility of steps at a large driving force.[2)] When high speed-steps collide, the steps pass through each other making another atomic layer on the facet due to the inertia of the liquid accompanying the step motion. This kinematical multiplication of steps makes the step density higher and the growth of the facet faster. Mobility of steps for a ^{4}He crystal in the superfluid liquid is not limited by the diffusion and can be very high at very low temperatures.[3)] We observed anomalously fast growth of the c-facet of a ^{4}He crystal induced by acoustic waves. The fast growth could not be explained by the spiral growth mechanisms and we possibly observed the multiplication of steps.

We reported that acoustic waves induce crystallization of ^{4}He crystals at low temperatures.[4,5)] We interpreted that this interface motion was induced by the acoustic radiation pressure. For the small displacement of the interface by a short acoustic wave pulse (1 ms), the acoustic radiation pressure model can explain the growth velocity of the rough and vicinal surfaces reasonably well at low temperatures by taking into account the orientation dependence of the growth coefficients. In this paper, we report the effect of the longer pulse (50 ms) applied to the vicinal surface from the crystal side. The displacement of the interface was much larger and the clear c-facet soon appeared on the top of the upheaval because the growth velocity of the c-facet was smaller than the vicinal surface. This growth velocity of the c-facet was much larger than the value expected from the spiral growth model with known step mobility. Since the pressure oscillation of the applied acoustic wave was large enough to accelerate steps to the velocity of sound, kinematical step multiplication was likely to occur. We constructed a growth model of the facet with this multiplication process and found a reasonable agreement with the experiment.

Experiments were performed in the optical cryostat described in our previous papers and a ^{4}He crystal inside a sample cell was observable from room temperature.[4,5)] We had two ultrasound transducers of $LiNbO_3$ in the cell facing each other 10 mm apart. The frequency of the acoustic waves was 9 MHz and their directions were vertical. The effective diameter of the transducer was about 5 mm. We used ^{4}He of regular purity; ^{3}He concentration was of the order of 0.1 ppm. It was too small to produce a noticable effect on the step mobility.[3)] We nucleated a ^{4}He crystal on the upper transducer from metastable superfluid ^{4}He with an overpressure of a few mbar using an acoustic wave pulse.[6)] As the seed crystal grew, it sometimes fell on the lower transducer with its facet parallel to the transducer surface. By this procedure, we were able to obtain the oriented crystal within several trials. The crystal was grown to fill the lower part of the sample cell and the position of its interface was adjusted between the two transducers so as to apply acoustic waves upwards from the lower transducer. Interface motion or crystallization induced by the acoustic waves from the crystal side was recorded using a high-speed camera synchronized with the acoustic wave pulse. Growth velocity v was measured as a function of temperature T and acoustic wave power density I. I is related to the chemical potential difference between the solid and the liquid $\Delta\mu$ or the driving force for the crystallization as

$$I = \frac{\rho c}{2}\Delta\mu \tag{1}$$

from the acoustic radiation pressure model.[5)]

Calibration of I is important in order to compare the experimental results with growth models. We applied a long acoustic wave pulse to a crystal–superfluid interface, and the interface above the transducer moved upwards with the displacement h brought about by acoustic radiation pressure and stopped at the position where the force balanced with gravity. The balancing condition is given as

$$(\rho_c - \rho_l)gh + \frac{2\alpha}{R} = \frac{2I}{c_c}, \tag{2}$$

where ρ_c, ρ_l, g, α, R, and c_c are density of the crystal, density

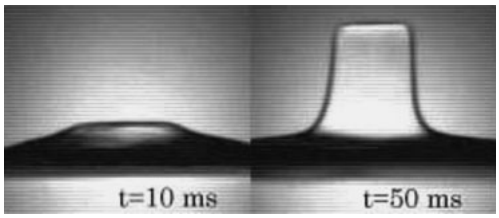

Fig. 1. Growth shape of c-facet taken by high-speed camera at $T = 200$ mK for $\Delta\mu = 59\,\mathrm{cm}^2/\mathrm{s}^2$. The pulse duration was 50 ms. The diameter of the c-facet was 3 mm at $t = 50$ ms.

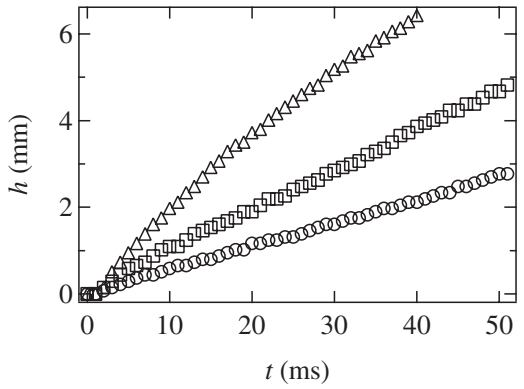

Fig. 2. Height of c-facet during application of acoustic wave pulses at $T = 100$ mK for $\Delta\mu = 38$ (circles), 59 (squares), and 90 (triangles) $\mathrm{cm}^2/\mathrm{s}^2$.

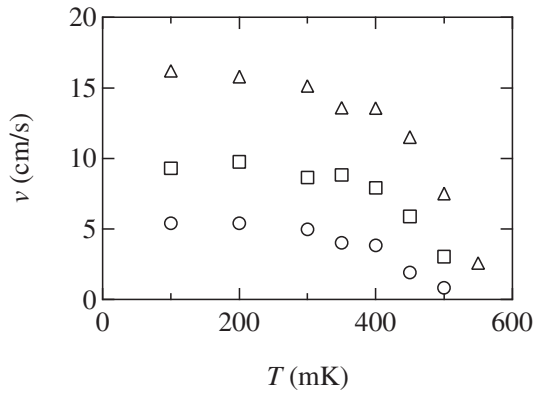

Fig. 3. Growth velocity of c-facet as function of temperature for $\Delta\mu = 38$ (circles), 59 (squares), and 90 (triangles) $\mathrm{cm}^2/\mathrm{s}^2$.

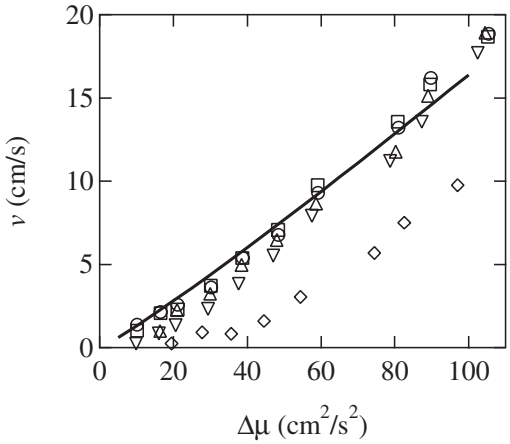

Fig. 4. Growth velocity of c-facet against $\Delta\mu$ at $T = 100$ (circles), 200 (squares), 300 (up triangles), 400 (down triangles), and 500 (diamonds) mK. The solid line is a plot of eq. (10).

of the liquid, the gravitational acceleration, the surface stiffness, the curvature radius of the interface, and the velocity of sound in the crystal. I in this procedure was about 2.5 times larger than the calibration adopted in our previous papers.[4,5] In the previous calibration we used a superfluid free surface and neglected the contribution from surface tension. We believe the calibration using the crystal–superfluid interface is more reliable and adopt this calibrated value in the following sections.

In Fig. 1, a typical crystal growth shape during application of the acoustic wave pulse is shown. The initial interface was the vicinal surface tilted from the c-facet by about 5° and was horizontally flat without an acoustic wave. The interface above the transducer moved upwards when the acoustic wave was applied for 50 ms. In this case, $T = 200$ mK and $\Delta\mu = 59\,\mathrm{cm}^2/\mathrm{s}^2$. The clear c-facet appeared on the top of the upheaval and its diameter was about 3 mm. The height of the facet h is plotted in Fig. 2 as a function of time t during the acoustic wave pulse at $T = 100$ mK for several values of $\Delta\mu$: $\Delta\mu = 38$ (circles), 59 (squares), and 90 (triangles) $\mathrm{cm}^2/\mathrm{s}^2$. We can obtain the growth velocity of the c-facet v by the linear fitting of $h(t)$. As can be seen in Fig. 2, the initial slope of $h(t)$ was slightly steeper and we excluded these data for the fitting. The initial fast growth is presumably the growth of the original vicinal surface existing before the c-facet appeared on the top. If we applied a much longer pulse, $h(t)$ eventually reached the saturation value as explained in the previous section.

We plotted the temperature dependence of v in Fig. 3 for $\Delta\mu = 38$ (circles), 59 (squares), and 90 (triangles) $\mathrm{cm}^2/\mathrm{s}^2$. Growth was faster for a larger I. It was faster at a lower T but almost saturated below 400 mK. The power dependence of v is shown in Fig. 4 and v increased linearly to $\Delta\mu$ below 400 mK. We can also see the saturation of v at low temperatures in this figure.

Although this growth velocity was slower than that of the vicinal surface of 5°, v was much larger than expected from the spiral growth mechanism as explained in the next section. For the spiral growth mechanism, $v \propto \Delta\mu^2$ and v should increase at a lower T without saturation. These features are not consistent with the observation and thus a new growth mechanism is needed to explain the experimental results.

Consider the growth of a c-facet of a ^{4}He crystal induced by a high-amplitude acoustic wave of the form $\sin(\omega t)$. According to the concept of acoustic radiation pressure,[7] the effective driving force on the crystal–liquid interface consists of two terms, the first- and second-order terms:

$$\delta\mu = \delta\mu_1 \cdot \sin(\omega t) + \delta\mu_2, \tag{3}$$

where the first-order term is related to the pressure amplitude in the wave δP as $\delta\mu_1 = [(\rho_c - \rho_l)/\rho_c\rho_l]\delta P$, and the second-order term is a function of time with a nonzero average: $\langle\delta\mu_2(t)\rangle = \Delta\mu$.

The value of the second-order term depends on the reflection coefficient at the interface R, which in turn depends on the interface mobility, and $R = 1$ if the mobility

is high.[8,9] Generally, the facet mobility is very low compared with the mobility of a rough surface, and the condition $R = 1$ cannot be justified in application to the whole facet. However, here we should take into account that the growth of a facet is only due to the motion of elementary steps. In this case, the concept of radiation pressure should be applied directly to the step motion. At low temperatures, the mobility of steps is known to be very high.[10] Therefore, effectively, the condition $R = 1$ still holds, and one can write an equation similar to that in our previous paper:[4,5]

$$\Delta\mu = 2\left(\frac{\delta P}{\rho_c c_1}\right)^2. \tag{4}$$

Usually a facet grows via the motion of elementary steps, which form spirals rotating around screw dislocations terminated at the facet. The growth velocity v is proportional to the density of steps n_s and their velocity v_s. Under a small driving force $\delta\mu$, both are proportional to $\delta\mu$, and v can be written as[1]

$$v \cong \frac{\rho_c \eta}{19\beta}(a \cdot \delta\mu)^2, \tag{5}$$

where β and a are the step energy (per unit length) and step height, respectively, and η is the step mobility defined as $\eta = v_s/\delta\mu$.

A driving force of the form of eq. (3) produces periodic growth and melting, and the net growth velocity of the facet is only due to the second-order term in this equation. This velocity is very low, 3–4 orders of magnitude lower than those observed in the present work. For instance, with $\beta/a = 0.011\,\mathrm{erg/cm^2}$ and $\eta = 20\,\mathrm{s/cm}$, which corresponds to $T = 200\,\mathrm{mK}$[10,11] at $\Delta\mu = 50\,\mathrm{cm^2/s^2}$, eq. (5) gives $v = 1.5 \times 10^{-3}\,\mathrm{cm/s}$.

The observed high growth velocities of facets also cannot be the result of two-dimensional (2D) nucleation. First, in this case one would expect an opposite temperature dependence of the growth speed and second, a quite different dependence on the driving force. Numerical estimates, based on standard formulas for 2D nucleation,[3] also give an excessively low growth speed even at the highest explored temperatures and driving forces.

Thus, conventional growth mechanisms fail to explain our observations. Here, we present an explanation based on the idea of kinematic multiplication of elementary steps. According to Parshin and Tsymbalenko,[2] at low temperatures and under strong driving forces the dynamics of steps on the surface of helium crystals becomes essentially nonlinear. In particular, n_s can significantly increase via the mechanism of kinematic multiplication. At high velocities, $v_s > v_0 \approx 0.3c$ (c is the velocity of sound in the liquid), a new process becomes possible: overthrow of steps into the next atomic layer at the collision of steps of opposite sign. Thus, the created seed of the new atomic layer starts to grow if its initial size exceeds the critical radius, $R_c = \beta/a\rho_c|\delta\mu|$; otherwise it shrinks and eventually collapses. In the process of growth, the seed meets another seed; their boundaries (step loops) collide, a new seed is created and the process continues. Such a multiplication of steps and seeds of new atomic layers can result in continuous growth of the whole facet that is much faster than usual spiral growth.

In order to trigger this growth mechanism, a few preexisting steps (associated with screw dislocations, for instance) are necessary; to reach v_0 and even higher speeds, the step mobility η should be sufficiently high,

$$\eta \gg \frac{v_0}{|\delta\mu|}. \tag{6}$$

Furthermore, the acceleration time from zero velocity to v_0 should be very small compared with the period of oscillations, i.e.,

$$\omega \ll \frac{a\rho_c|\delta\mu|}{m v_0}, \tag{7}$$

where m is the step effective mass (per unit length), which is estimated as $m \approx 5 \times 10^{-18}\,\mathrm{g/cm}$.[10]

Under these conditions, the number of step loops will progressively increase until it reaches a maximum determined by geometrical limitations. Namely, the maximum possible number of loops of a given diameter d (per unit area) is estimated as $n_d \approx 1/d^2$, where d varies from $2R_c$ to the facet size D. Then the total *length* of steps per unit area L is

$$L \approx \sum_d \frac{\pi d}{d^2} \approx \frac{\pi}{2R_c}\log\frac{D}{R_c}. \tag{8}$$

The growth (or melting) speed of the facet can be written as $v = aLv_s$, where positive (negative) v and v_s correspond to growth (melting). With eq. (8), we obtain

$$v \approx \frac{\pi a v_s}{2R_c}\log\frac{D}{R_c} = \frac{\pi a^2 |v_s|\rho_c\delta\mu}{2\beta}\log\frac{D}{R_c}. \tag{9}$$

On the right-hand side of eq. (9), it is assumed that contributions of steps of different length to L follow variations of $\delta\mu$ in time at all length scales, up to D, which was about 3 mm in our experiments (see Fig. 1). In fact, it could be true only at scales up to l, the characteristic displacement of steps during the period of oscillations, $l \approx v_s/\omega$. At larger scales, the distribution of steps basically remains unchanged and does not contribute to average growth velocity. Also we should take into account that with condition (6) v_s is limited by some critical velocity, presumably by the velocity of sound in the liquid c. Therefore, we set $v_s = \mathrm{const} = c$ and substitute D by c/ω ($10^{-3}\,\mathrm{cm}$) while averaging eq. (9) in time. Finally, we have for the average growth velocity:

$$v \approx \frac{\pi a^2 c\rho_c\Delta\mu}{2\beta}\log\frac{c}{\omega R_c}, \tag{10}$$

where average R_c is estimated as $R_c = \beta/a\rho_c\delta\mu_1$. $R_c \approx 3 \times 10^{-6}\,\mathrm{cm}$ at $\Delta\mu = 100\,\mathrm{cm^2/s^2}$. We see that in this regime the growth velocity changes almost linearly with $\Delta\mu$ and does not depend on temperature, in agreement with experimental observations. Numerically, eq. (10), without any adjustable parameter, fits the experimental data well as shown by the solid line in Fig. 4.

The mobility of a vicinal surface could be considered in a similar approach. The main difference is due to the presence of a number of linear steps, which may significantly contribute to the growth velocity. However, in order to estimate this contribution, one should take into account the dependence of v_s in eq. (9) on $\delta\mu$, which clearly should exist even in a supercritical regime ($v_s > c$). Note in this connection that the account for this dependence should also

somewhat increase the growth velocity of a facet compared to eq. (10).

It is easy to confirm that condition (7) is valid at all values of acoustic amplitudes used in the experiment. As for condition (6), it is valid at all temperatures below 400 mK. However, due to the fast drop of the step mobility η, at 400 mK ($\eta \cong 2$ s/cm, see ref. 11) and especially at 500 mK ($\eta \cong 1$ s/cm) this is not the case any more. At $\Delta\mu \cong 25$ cm^2/s^2, we have $\delta\mu_1 \cong 1 \times 10^4$ cm^2/s^2 and thus $\eta\delta\mu_1/c \cong 1$ at $T = 400$ mK. At $\Delta\mu \cong 100$ cm^2/s^2, $\eta\delta\mu_1/c \cong 1$ at $T = 500$ mK. This explains the observed decrease in v at 400–500 mK. Note also that at $T = 500$ mK and $\Delta\mu$ below 15–20 cm^2/s^2 the growth velocity is practically zero, which means that the discussed mechanism indeed does not work any more, if the step velocities fall below $0.3-0.5c$.

We observed very fast growth of the c-facet of a ^{4}He crystal induced by acoustic waves. The growth velocity cannot be explained by the spiral growth model for the known values of the step mobility. We developed a new mechanism for crystal facet growth by application of high-power acoustic waves. Considering the first-order effect of the oscillating pressure of the acoustic wave, steps are accelerated to the order of the sound velocity. Collisions of such high-speed steps can induce the multiplication of steps as Parshin and Tsymbalenko suggested[2)] and lead to a higher step density than that of the spiral growth. This model reproduces the observed power and temperature dependences well and also quantitatively agrees with the experiment. Thus, our observations may be considered as a first manifestation of the effect of kinematic multiplication of elementary steps on a crystal surface.

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