Elementary Steps on the ^{4}He Crystal Interface Probed by ^{3}He Atoms

V. Tsepelin, J. P. Saramäki, A. V. Babkin,[*] P. J. Hakonen, J. J. Hyvönen, R. M. Luusalo,
A. Ya. Parshin,[†] and G. K. Tvalashvili[‡]
Low Temperature Laboratory, Helsinki University of Technology, 02150 Espoo, Finland
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The growth dynamics of **c** facets, governed by individual Frank-Read type of sources, has been studied at millikelvin temperatures in the presence of small concentrations of ^{3}He atoms in the liquid. We find that in the spiral growth regime, interaction of ^{3}He atoms with moving elementary steps on the surface results in additional friction. Our results are compared with theory, where high-frequency ($\omega \geq T/\hbar$) zero-point oscillations of the steps are taken into account. There is a good agreement if we assume the amplitude of these oscillations $\xi \approx 5$ nm.

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The interface between superfluid and solid ^{4}He exists down to $T = 0$. In the low temperature limit where dissipation vanishes, quantum fluctuations become important for an adequate description of its kinetic properties. As an example the growth rate of facets of a ^{4}He crystal in the presence of ^{3}He impurities is predicted to depend strongly on the magnitude of the quantum fluctuations of steps [1]. Here we present the first experimental evidence of this phenomenon.

Crystal growth at low temperatures is usually assigned to the movement of steps. In the case of flat facets corresponding to high-symmetry orientations, dislocation lines ending on the surface create steps that wind around in a spiral staircase pattern in response to a large enough pressure difference across the interface. The growth of a faceted surface is thus governed by the forces acting on the steps, in particular by the friction caused by collisions of the moving step with thermal excitations and impurities in the liquid phase. The growth kinetics of ^{4}He crystals at low temperatures has been measured in detail for rough surfaces [2], vicinal surfaces [3], and facets [4]; theoretical discussion has been presented in Refs. [1,5,6]. In our previous work [4], the growth of **c** facets was investigated using interferometric methods. In the work presented in this Letter, we have studied the effect of small amounts of ^{3}He atoms on the spiral growth of **c** facets of ^{4}He crystals at temperatures $10 \leq T \leq 200$ mK.

At low enough temperatures ($T \leq 100$ mK) collisions of steps with ^{3}He atoms in the bulk liquid are expected to contribute significantly to the growth resistance of the facet [1], in addition to collisions with phonons dominant at higher temperatures [3,5]. Within our temperature and ^{3}He concentration range the magnitude of this effect is expected to depend on the effective width of the elementary steps, which affects the scattering process. Our results are in agreement with this picture—a decrease of the step mobility with increasing ^{3}He concentration in the liquid was clearly observed at $T \leq 100$ mK, in addition to the concentration-independent mobility at higher temperatures. The obtained mobilities are in accord with the theory presented in Ref. [1], which stresses the role of high-frequency zero-point oscillations of steps.

In our experiments the crystals were grown inside an optical cell monitored by a two-beam interferometer, similar to the one used in our previous work [4]. The experimental chamber is schematically illustrated in the inset of Fig. 1. The body of the chamber is a polished copper cylinder ($\phi = 19$ mm, $h = 15$ mm), sealed from the top and bottom by two fused-silica windows. The antireflection-coated bottom window acts as our optical reference plane (RP). The interference patterns created by guiding laser light ($\lambda = 632.8$ nm) to the cell were viewed with a cooled CCD camera [4]. The cell was thermally connected to the mixing chamber of a dilution refrigerator, and the temperature was measured with a carbon resistor.

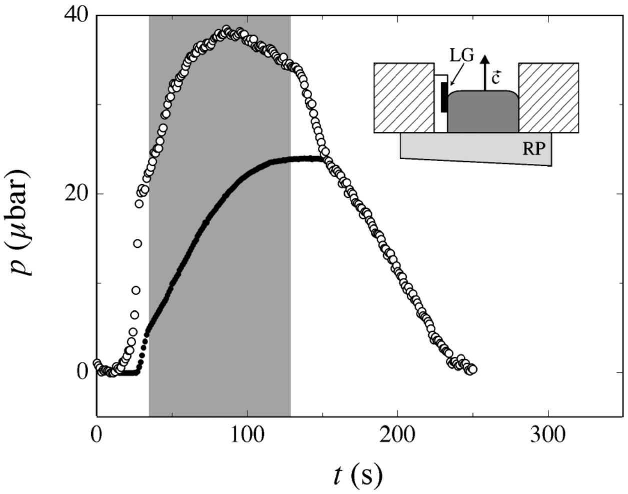

FIG. 1. Illustration of the liquid pressure (open circles), measured by the pressure gauge, and the hydrostatic variation at the contact point of the capacitor and the interface, calculated from the level gauge data, during one cycle of growth and melting. In the beginning of the cycle, the crystal shape changes as the contact point of the rough surface at the capacitor moves closer to the facet level. Thereafter, it follows the movement of the facet (shaded area). The inset shows a schematic drawing of the experimental cell.

The crystals were nucleated from liquid helium at $T \simeq 20$ mK by applying a high voltage to a capacitive nucleator similar to the one described in Ref. [7]. The two-beam interferometer was used to select a crystal with the correct alignment, having the **c** facet horizontal with an accuracy of 10^{-4} rad [4,8]. Initially, the crystal was grown from regular commercial helium with nominal ^{3}He concentration of less than 100 ppb. Then small amounts of ^{3}He were gradually added to the liquid using calibrated ballast volumes at room temperature and driving helium in and out of the cell by connecting the volumes to the sinusoidally heated ballast described below. Before adding ^{3}He, the crystal was melted to a small size.

The level of the superfluid/solid interface was measured by an interdigital capacitor mounted on the cell wall (LG). The capacitor was produced by evaporating a thin layer of chromium in the shape of a vertically aligned fingered structure on fused silica glass (finger width 10 μm, spacing 10 μm). The capacitance of the level gauge was measured with a capacitance bridge, consisting of an ac voltage source, a ratio transformer, and a lock-in-amplifier. The liquid pressure was monitored with a Straty-Adams type pressure gauge [9], connected to a high-resolution Andeen-Hagerling 2500 A capacitance bridge. The pressure gauge was calibrated against the level gauge by measuring the change in the hydrostatic pressure (resolution ≈0.3 μbar) when slowly growing and melting the crystal over the whole 4.8 mm level gauge range.

The crystal was periodically grown and melted by applying a sinusoidal voltage to a heater wire wound around a 100 cm^3 ballast volume, connected to the helium fill line outside the cryostat. Frequencies between 2–8 mHz were used for this purpose. While the crystal was growing and melting, the level of the liquid/solid interface and the liquid pressure were simultaneously measured. Highest measured **c**-facet velocities were slightly below 50 μm/s.

The **c** facet fills only the central part of the cell; it is the adjacent, curved rough surface that touches the capacitor. Thus the level gauge signal represents the movement of the rough surface. Figure 1 illustrates the total cell pressure measured by the Straty-Adams gauge and the hydrostatic variation calculated from the level gauge data during an oscillation cycle. In the beginning of the cycle, once a threshold pressure is exceeded, the round part of the surface touching the wall changes its shape rapidly, and the contact point to the capacitor moves up ≈0.25 mm, approaching the facet level. After this, the motion of the rough surface corresponds quite closely to the movement of the facet—some small difference is still expected, but may be considered as negligible. When the crystal is melting, the round part lags behind its equilibrium shape.

All our crystals showed a growth threshold $\Delta p_0 = p_0 - p_{\mathrm{eq}}$ with respect to the equilibrium melting pressure p_{eq}. For different crystals the values of Δp_0 were scattered within relatively wide range 1–25 μbar, from where the corresponding size of a dominating growth source $r \propto 1/\Delta p_0$ can be estimated as 1–0.05 mm. This must be a Frank-Read source [10] or a single screw dislocation located close to the facet edge. In both cases the growth velocity of a facet at $p > p_0$, immediately above the threshold, behaves as $v \propto \sqrt{p - p_0}$. In the case of a Frank-Read source we obtain

$$v_1 = \frac{4\sqrt{2}\, a^2 \Delta p_0^2 \mu}{\pi^2 \beta} \left(\frac{\Delta\rho}{\rho_L} \right)^2 \sqrt{\frac{p - p_0}{\Delta p_0}}\,, \tag{1}$$

where $\Delta\rho = \rho_S - \rho_L$ is the difference in densities of the solid and liquid phases of helium, whereas for a single dislocation this velocity is less by a factor of $\sqrt{2}$ (with the assumption that the distance of the dislocation from the facet edge does not depend on p_0). Far from the threshold we have a usual quadratic dependence $v \propto \Delta p^2$ [6,11], where $\Delta p = p - p_{\mathrm{eq}}$ (dotted line in Fig. 2)

$$v_2 = \frac{\Lambda \mu a^2}{\pi \beta} \left(\frac{\Delta\rho}{\rho_L} \right)^2 \Delta p^2, \qquad \Lambda = 0.330\,95\ldots. \tag{2}$$

In order to describe the experimental data in the whole range of overpressures we used a simple interpolation formula $1/v = 1/v_1 + 1/v_2$ (solid line in Fig. 2).

From data similar to those presented in Fig. 1, v vs Δp relationships were calculated for each concentration and temperature, averaging over several series of measurements. These measurements were performed at $10 \le T \le 225$ mK, with 40, 110, and 220 ppm of ^{3}He in the liquid phase (estimated error ±20 ppm). Figure 3 shows the measured facet velocities v as a function of overpressure Δp at different concentrations of ^{3}He. This velocity is related to the step velocity v_s as $v = (a/d)v_s$, where $a = 3 \times 10^{-8}$ cm is the lattice spacing and d is the separation of the spiral arms. The solid lines in Fig. 3 indicate fits of our interpolation formula, with the step mobility μ and the threshold value p_0 as the fit parameters, showing good agreement with the measured data. The classical picture may be considered adequate

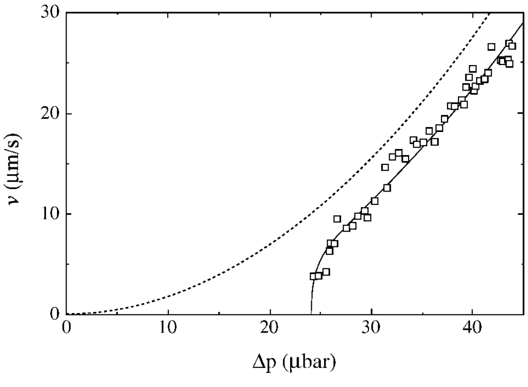

FIG. 2. v vs Δp dependence for one of the samples with $n_3 = 110$ ppm in the liquid phase measured at $T = 50$ mK. Growth threshold $p_0 \simeq 24$ μbar corresponds to the size of the growth source $\simeq 0.05$ mm. Lines are explained in the text.

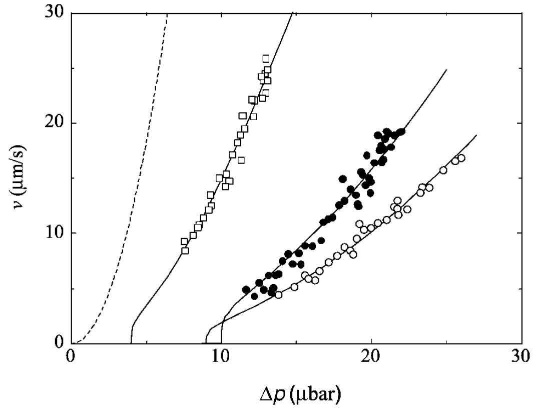

FIG. 3. Growth velocities measured at $T = 37$ mK with concentration 20 ppm (∘). The solid lines are the best numerical fits of the Eq. (3), using μ and p_0 as free parameters. The dashed line corresponds to pure ^{4}He (0.1 ppm).

within our range of velocities and driving pressures due to the following facts: (1) For most of our data, the velocities are not very high, of the order of 10 m/s. Therefore, the kinetic energy of the step is much less than the step energy $\beta = 0.011$ erg/cm^2 [3], and thus the effective mass of the step does not have to be taken into account as in the modified theory of spiral growth [4]. (2) Since collisions of the steps with ^{3}He atoms provide an additional means for energy dissipation, localization effects are expected to be of importance only at rather high Δp's.

Figure 4 displays the obtained step mobilities as a function of temperature for different ^{3}He concentrations. As the concentration of ^{3}He atoms is increased, there is a clear decrease in the mobilities at low temperatures, in accord with the theory presented in Ref. [1]. There are two mechanisms by which the added ^{3}He atoms may affect the step mobility: the ^{3}He atoms in the liquid phase behave as a 3D gas, scattering at the steps, whereas the ^{3}He atoms adsorbed at the interface may be considered as an in-plane 2D Fermi gas of quasiparticles [12,13]. At our temperature and concentration range, the 2D gas is expected to be saturated. The amount of ^{3}He atoms dissolved in the solid phase is negligible at temperatures well below 0.3 K [14]. The mobility is thus determined by the friction due to step collisions with phonons and with ^{3}He atoms in the bulk liquid. At temperatures $T \geq 100$ mK, collisions of the steps with phonons dominate; the corresponding growth resistance is proportional to T^3 [3]. At $T < 100$ mK the phonon resistance becomes very small, and the scattering of impurities plays the major role.

According to Ref. [1], it is the relation between the effective width of the step and the impurity wavelength that determines the scattering amplitude and consequently the step mobility. The scattering can be assumed to be elastic since at low temperatures the step is effectively rigid, meaning that the typical wavelength of the fluctuations in the step position x is larger than the impurity wavelength. Further, the scattering process appears to be rather slow: a ^{3}He particle stays near the step a time $\tau \sim \hbar/T$. Thus in the presence of high-frequency ($\omega \geq T/\hbar$) zero-point fluctuations in x, the incident particles "see" only an average profile of the step. Note that for $T \leq 200$ mK the effect of thermal fluctuations of the step is small and may be neglected.

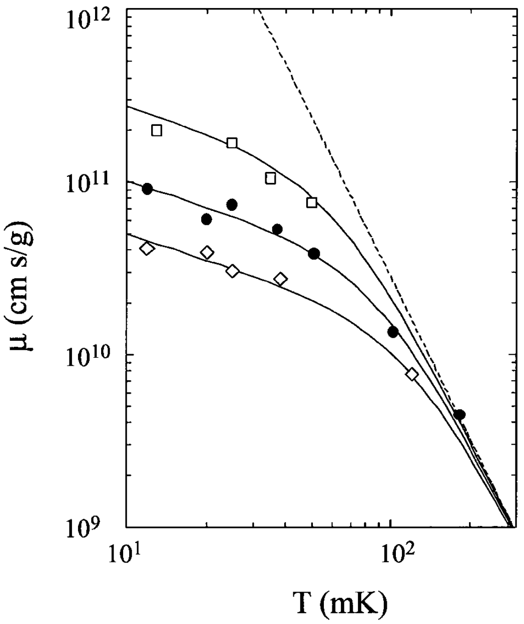

FIG. 4. Step mobilities μ calculated from the measurements with ^{3}He concentrations of 40 ppm (□), 110 ppm (•), and 220 ppm (◇). Earlier data, measured with regular ^{4}He (purity: 0.1 ppm) by Ruutu *et al.* are represented by the dashed line. The solid lines denote theoretical curves, calculated using $\xi = 5$ nm (see text for details).

In Ref. [1], the average step profile is an error function

$$z(x) = \frac{a}{\sqrt{2\pi\xi^2}} \int_{-\infty}^{x} \exp\left(-\frac{t^2}{2\xi^2}\right) dt \qquad (3)$$

with root mean square displacement ξ, which, depending on the temperature, is predicted to stay in the range 2.5–3 nm. If, following [3], the effective width of the step w is defined as the average distance on which the surface height goes from $0.1a$ to $0.9a$, then we find $w \simeq 3.3\xi \simeq 9$ nm. It is important to note that the theoretical model [1] suggests significantly larger values of the average amplitude ξ and the effective width w as compared to the correlation length $\xi_0 \simeq 0.6$ nm and the effective width of the step $w_0 \approx 2.4$ nm, which can be deduced from the experiments of the Paris group [3] in the frame of the "weak coupling" approximation [3,5].

Assuming $\xi \gg \xi_0$, within our range of temperatures ($T \geq 10$ mK) and low ^{3}He concentrations ($n_3 \leq 220$ ppm), the gas of impurities remains nondegenerate, and the contribution to the growth resistance due to the ^{3}He scattering is predicted to be

$$\frac{1}{\mu_3} = \frac{\sqrt{2}}{\pi} \frac{a^2\sqrt{m_3 T}}{\xi} n_3\,, \qquad (4)$$

where m_3 and n_3 are the effective mass and the concentration of ^{3}He atoms in the liquid, respectively. The total growth resistance containing both mechanisms is the sum, $1/\mu = 1/\mu_3 + 1/\mu_{\mathrm{ph}}$.

The solid lines in Fig. 4 were calculated by fitting the ^{3}He collision mobilities to the data, and summing the respective contributions to the growth resistance. The phonon limited mobility was fitted to the high temperature data as $1/\mu_{\mathrm{ph}} = AT^3$, where A is a fit parameter, yielding $A = 10^{-7.5}\ \mathrm{g\,s^{-1}\,cm^{-1}\,K^{-3}}$. The value of ξ was estimated by fitting Eq. (4) to the mobility data at temperatures $T \leq 100$ mK, resulting in $\xi = 5 \pm 2$ nm. We see that experiments show even wider steps than it was estimated above. Furthermore, in the phonon collision regime a good agreement is found with the data measured earlier by Ruutu *et al.* [4]. When discussing the difference between the calculated and measured values of ξ, one should keep in mind that the calculations were made using a simple harmonic approximation, which is not expected to be very accurate in the case of large fluctuation amplitudes. From this point of view, the measured value of ξ rather than the calculated one should be considered as a new important characteristic of the step structure.

In conclusion, we have shown that the growth dynamics of the **c** facets of ^{4}He crystals is significantly affected by the presence of ^{3}He atoms in the bulk liquid. Our data support the physical picture presented in [1], where the step is considered as a quantum string with large amplitude of zero-point oscillations. The step-impurity interaction results in additional friction, which is described with one new parameter, the root mean square displacement ξ. The experimental value $\xi = 5 \pm 2$ nm is large compared to the interatomic distance, and does not depend on the ^{3}He concentration. In fact, its value is even larger than the effective width measured by the Paris group [3]. This means that the coupling of the interface with the lattice might be even weaker than it was previously thought.

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*Corresponding author.
Present address: Department of Physics and Astronomy, University of New Mexico, 800 Yale Blvd., Albuquerque, NM 87131.

†Permanent address: P. L. Kapitza Institute for Physical Problems, ul. Kosygina 2, 117334, Moscow, Russia.

‡Present address: Department of Physics, Lancaster University, Lancaster LA1 4YB, UK.

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