

## Anisotropy of Growth Kinetics of $^3\text{He}$ Crystals below 1 mK

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The growth anisotropy of different facets has been measured in  $^3\text{He}$  crystals at 0.55 mK using a low-temperature Fabry-Pérot interferometer and high-resolution pressure measurements. The observed linear dependence of the growth velocity on the driving force shows that facets grow due to the presence of dislocations. The values of the obtained step energies suggest that  $^3\text{He}$  has stronger coupling of the liquid-solid interface to the lattice than has been expected. The dependence of the step energy versus the step height is consistent with a quartic power law pointing out that the step-step interactions are of elastic origin.

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The interface between the superfluid and solid phases of helium offers a unique opportunity to study crystal growth processes and the underlying physics of the basic excitations on the interface. This is because the vanishing latent heat of crystallization and good thermal conductivities of both bulk phases yield fast dynamics at low enough temperatures. Thus the thermal processes as well as the mass flows affect, under these conditions, the crystal growth much less than in a case of a typical liquid-solid interface.

Up to now extensive surface studies have been carried out with  $^4\text{He}$  crystals. The growth properties of rough and smooth surfaces (facets) have been investigated both experimentally and theoretically [1–3]. On rough surfaces of crystals, growth is easy because of the abundance of sticking sites for atoms to be added to the solid phase. On smooth facets, corresponding to high symmetry crystallographic orientations, the growth rates are significantly lower. Facet growth is usually assigned to the presence of dislocations which end on the surface of the crystal and form elementary steps. The two surface states, rough and smooth, are separated by a so-called roughening transition [4,5].

The recent findings of Tsepelin *et al.* [6,7], who observed a total of eleven different types of facets on growing  $^3\text{He}$  crystals, make  $^3\text{He}$  actually even more interesting than  $^4\text{He}$  from a morphological point of view. With so many different facets the growth anisotropy of  $^3\text{He}$  crystals could be mapped out and this would give direct information about the interaction of the liquid-solid interface with surface excitations. So far only an average growth rate of  $^3\text{He}$  crystals has been reported [8–10].

In order to reach the regime where the mobility of the superfluid-solid interface of  $^3\text{He}$  is not limited by bulk thermal impedances, very low temperatures are needed. The measurements have to be conducted well below the Néel temperature,  $T_N = 0.93$  mK [11], at which the spins of solid  $^3\text{He}$  order antiferromagnetically.

In this Letter we present the results of our interferometric experiments in which we have been able to measure, for the first time, the growth anisotropy of  $^3\text{He}$  crystals at a temperature of 0.55 mK. The normal velocities of ten

different types of facets were measured and the energies of elementary steps on these facets were calculated.

Our experimental setup is described in Ref. [12]. Single  $^3\text{He}$  crystals were grown in a Pomeranchuk-type cell. The crystals were imaged with a low-temperature Fabry-Pérot interferometer and the growth velocities of facets were determined by tracking the positions of interference fringes between the subsequent interferograms.

The experimental data were obtained in the following manner: after melting the crystal to get a rounded (rough) interface, the pressure of  $^3\text{He}$  was slowly increased, while every 4 s an interferogram was recorded. Because the facets with high Miller indices grow quite fast, these are only seen at rather small overpressures. After several minutes the overpressure was increased in order to measure the growth rates of the more stable, slowly growing facets. Data of the various facets were used only when there were at least three equidistant and parallel interference fringes for a facet (to identify the facet unambiguously from the measured angles) and the facet was visible for at least five subsequent interferograms (typically for many more). Sometimes it was observed that the growth rate of a certain facet varied somewhat after the crystal had been subjected to an extended period of growth or to a period of rapid growth. While this does not affect our results qualitatively, we believe it gives better insight in the ratio of the growth velocities of the different facets to present and compare the data of one particular “growth run.” It means that Fig. 1 does not contain data on some of the facets observed in other growth series.

Figure 1 summarizes the measured growth rates  $v$  obtained for different types of facets during one continuous growth series as a function of overpressure  $\delta p$ , which is the pressure difference between the actual pressure and the equilibrium melting pressure at the measurement temperature of 0.55 mK. For (210) facets there are two sets of data points because two different facets of the same type exhibited somewhat different growth rates.

The observed growth displays a rather strong anisotropy: for instance, the measured velocities of the (110) and (510) facets differ by about 1 order of magnitude. The

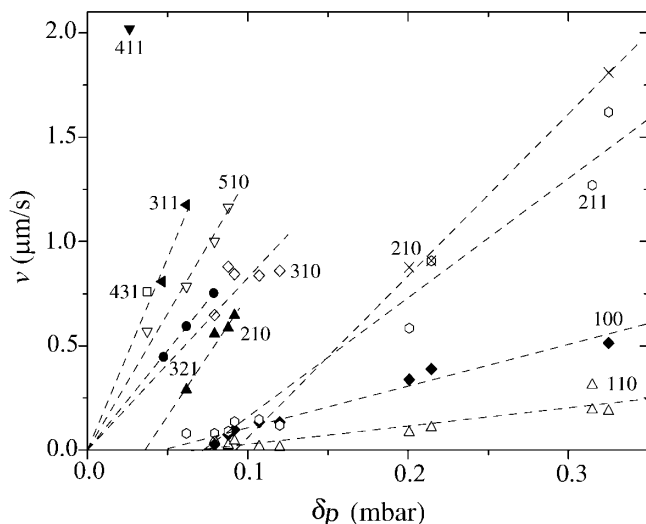


FIG. 1. Anisotropy of  $^3\text{He}$  crystal growth at  $T = 0.55$  mK. The dashed lines are the linear fits to the data for different facets marked with Miller indices.

increase of the growth velocity with the applied driving force  $\delta p$  shows a linear dependence and this is in agreement with measurements made at higher temperatures of (0.6–0.8) mK by Nomura *et al.* [9] and Akimoto *et al.* [10]. The values they obtain for the growth velocities of  $^3\text{He}$  crystals are similar to our results for the most stable facets (110) and (100), but this is the first time that the growth velocities can be assigned to particular facets.

The linear dependence of the growth velocity of a facet on the driving overpressure indicates that in our experiments growth cannot be due to thermal activation of two-dimensional terraces on a facet, since at 0.55 mK this would give growth rates many orders of magnitude below the measured values, and also a different dependence on  $\delta p$  [9,13]. Thus we conclude that the dominating growth mechanism is spiral growth which is the main growth mechanism in the presence of screw dislocations. Typically spiral growth occurs only for a driving force larger than a certain critical value, at which the step can escape from the Frank-Read sources [13]. Figure 1 shows that we have a maximum critical overpressure of about 0.05 mbar for the most stable facets, from which we conclude that this particular crystal could contain up to a few hundred dislocations per  $\text{mm}^2$ .

The growth rate  $v$  for spiral growth of a faceted liquid-solid interface shows a different dependence on the applied overpressure in two different growth regimes, of constant or suppressed step mobility [2,13]. In the regime of constant mobility  $v$  is given by

$$v = \frac{\mu d^3}{19\beta} \left( \frac{\rho_s - \rho_l}{\rho_l} \right)^2 K (\delta p)^2. \quad (1)$$

Here  $\beta$  is the step energy [14],  $\mu$  is the step mobility defined as the step velocity per unit driving force [2],  $d$  is the height of an elementary step on a facet,  $\rho_s$  and  $\rho_l$  are the densities of the solid and liquid phases, respectively,

and  $K$  is the number of steps produced by one dislocation [2,13].

The step mobility can be estimated in the following way. At temperatures below  $T_N$  in  $^3\text{He}$  there are two main dissipative mechanisms related to the step motion: scattering of magnons from the solid and quasiparticles from the liquid at their collisions with a moving step. The magnon contribution to the step resistivity,  $1/\mu_m$ , can be calculated using the known scattering cross section [15]. Assuming specular (100%) reflection at the interface and neglecting the anisotropy of the magnon velocity  $c$ , one obtains

$$1/\mu_m \approx \frac{(k_B T)^4 d^2}{w \hbar^3 c^4}, \quad (2)$$

where  $k_B$  is the Boltzmann constant,  $\hbar$  is the Planck constant, and  $w$  is an effective width of the step which is expected to be of the order of a few lattice constants  $a$  as in  $^4\text{He}$  crystals [3,16].

The scattering cross section of quasiparticles is difficult to calculate since their wavelength is very short,  $k_F a \sim 4$ , where  $k_F$  is a Fermi wave vector. As an order of magnitude this cross section can be estimated as  $\sigma \sim d^2/w$  [15]. It yields for the contribution  $1/\mu_{qp}$  due to quasiparticles

$$1/\mu_{qp} \sim \hbar k_F^4 \frac{d^2}{w} \exp(-\Delta/T), \quad (3)$$

where  $\Delta$  is the superfluid energy gap of  $^3\text{He-B}$ . This estimate is valid at low step velocities,  $v_s < k_B T / \hbar k_F$ ; at larger velocities  $1/\mu_{qp}$  decreases [17]. Note that both contributions to the step resistivity,  $1/\mu_m$  and  $1/\mu_{qp}$  depend rather strongly on temperature.

While Eq. (1) suggests a quadratic dependence of the growth rate on the driving force, this remains valid only until the step velocity  $v_s$  reaches a critical velocity  $v_c$  when the step mobility suddenly decreases. In this regime of suppressed step mobility [2,13]

$$v = \frac{v_c d^2}{2\pi\beta} \frac{(\rho_s - \rho_l)}{\rho_l} K \delta p, \quad (4)$$

and the growth speed of a facet becomes linear in  $\delta p$ , independent of the step mobility and weakly dependent on temperature. Here the step inertia has been neglected because that becomes important only when the step velocity is of the order of the sound velocity [2]. The lowest critical velocities in  $^3\text{He}$  are the magnon velocity  $c$  in the solid and the pair-breaking velocity  $v_{pb}$ . At low magnetic fields both are about 7 cm/s [18,19]. The mobility estimates given in Eqs. (2) and (3) show that our experimental conditions (see Fig. 1) correspond to the regime of suppressed step mobility. This is also strongly supported by our qualitative observation that there is only a weak temperature dependence of the growth rates, in agreement with earlier measurements at somewhat higher temperatures [9,10].

Table I gives the results for the step energy  $\beta/K$  obtained from linear fits to the data in Fig. 1 and Eq. (4)

TABLE I. Miller indices of the planes in the bcc lattice, the squared interplanar distance ratio with respect to the (110) facet, the energy of the elementary step  $\beta/K$  obtained from one particular growth series of a single  $^3\text{He}$  crystal, and the possible values of  $K$ , the number of steps produced by one dislocation. The (111) facet was observed but not enough data were obtained to determine its growth speed.

| Miller index | $(d_{110}/d_{hkl})^2$ | $\beta/K$ (erg/cm)    | $K$       |
|--------------|-----------------------|-----------------------|-----------|
| 110          | 1                     | $6.6 \times 10^{-10}$ | 1         |
| 100          | 2                     | $1.4 \times 10^{-10}$ | 1,2       |
| 211          | 3                     | $3.3 \times 10^{-11}$ | 1,2       |
| 310          | 5                     | $1.4 \times 10^{-11}$ | 1,2,3     |
| 111          | 6                     | ...                   | 1,2,3     |
| 321          | 7                     | $8.6 \times 10^{-12}$ | 1,2,3     |
| 411          | 9                     | $8 \times 10^{-13}$   | 1,2,3,4   |
| 210          | 10                    | $7.0 \times 10^{-12}$ | 1,2,3,4   |
| 210          | 10                    | $4.9 \times 10^{-12}$ | 1,2,3,4   |
| 510          | 13                    | $3.4 \times 10^{-12}$ | 1,2,3,5   |
| 431          | 13                    | $2.2 \times 10^{-12}$ | 1,3,4     |
| 311          | 22                    | $1.4 \times 10^{-12}$ | 1,2,3,5,6 |

with  $v_c = 7$  cm/s. Also presented is the classification of the planes in the bcc structure of solid  $^3\text{He}$  [20], together with the interplanar distance  $d_{hkl}$  (which is equal to the step height on the corresponding facet) with respect to the  $d_{110} = 0.307$  nm. The value of  $K$ , which depends on both the facet orientation and Burgers vector of a dislocation, is not known, but the possible values for each facet are given in the last column of Table I. We want to point out that Table I presents our experimental data from one particular growth run, also shown in Fig. 1. The results of other growth runs indicate that there can be an uncertainty of up to tens of percents in the absolute values of  $\beta$  for the most stable facets, which is manifested by the two different  $\beta$  values for the two different (210) facets.

If there are many dislocations,  $K$  could have all the possible values for each observed facet, and the growth rate of a facet will be determined by the most “active” dislocations, with the maximum value of  $K$  for that facet. On the other hand, in our experiments basically the most stable facets have been observed, which have the lowest growth velocities, hence also minimum  $K$  value. In our analysis we have assumed this more stable situation in which one dislocation produces a single step, and the resulting values of  $\beta$  are presented in Fig. 2.

The linear fit to the data [except for the (411) facet] in the log-log coordinates in Fig. 2 yields approximately a quartic power-law dependence of the step energy. One can show that this dependence may be the result of elastic step-step interaction which has  $r^{-2}$  behavior [21]. Corresponding calculations are similar to those made by Landau who considered only the case of van der Waals interaction between steps ( $r^{-3}$ ), and not the elastic ( $r^{-2}$ ) interaction [22]. Quantitatively the correct order of magnitude is obtained for the step energies despite the uncertainty in the strength of the elastic interaction. Strictly speaking, such calculations are supposed to be valid only when applied to so-called secondary steps, i.e., steps on a vicinal

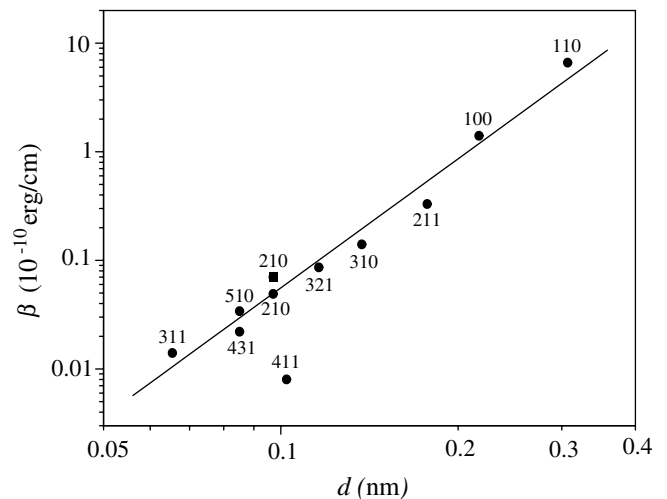


FIG. 2. Step energies  $\beta$  of different facets on a  $^3\text{He}$  crystal at  $T = 0.55$  mK for  $K = 1$ . The linear fit has a slope of  $3.95 \pm 0.25$ .

facet, where the distance between primary steps exceeds their effective width  $w$ , which is typically a few times the correlation length of the interface fluctuations  $\xi$  (see, e.g., Refs. [3,16]). For the case when  $w$  is larger than the distance between primary steps (weak coupling limit), an exponential dependence is expected rather than a power law [6]. Thus our observations indicate that, on  $^3\text{He}$  crystals,  $w$  is unexpectedly small, even smaller than on  $^4\text{He}$  crystals (see below). Note also that surprisingly the observed quartic power-law dependence extends not only to those facets which may be called vicinal, such as (510), but to the more closely packed lower order facets as well.

The energy of an elementary step on the (110) facet is  $\beta_{110} = 6.6 \times 10^{-10}$  erg/cm, which is unexpectedly larger than the value measured for the (1000) facet on  $^4\text{He}$  crystals [16]. According to Refs. [3,23] the width of an elementary step  $w$  is connected to the step energy as  $w\beta \sim \gamma d^2$ , where  $\gamma$  is the surface stiffness. Taking  $\gamma = 0.06$  erg/cm $^2$  [24], the calculated width of the step for the (110) facet,  $w_{110}$ , equals  $2a$ , which is about one-fourth of that in  $^4\text{He}$  [16]. The step width reflects the coupling strength of the interface to the crystal lattice and in  $^4\text{He}$  the step width of approximately  $10a$  has been attributed to the rather weak coupling [3]. The smaller step width in  $^3\text{He}$  crystals suggests that the coupling of the interface to the crystal lattice is strong in  $^3\text{He}$  compared with  $^4\text{He}$ .

The growth velocity of the rough surface was estimated in our experiments in a single observation when a macroscopic step propagated along the (100) facet at a temperature of 0.55 mK. The step width was about 3 mm and the analysis showed that, while only the (100) facet was growing, it gained about 60  $\mu\text{m}$  of height within 8 s under an overpressure of  $\Delta p = 65$   $\mu\text{bar}$ . The calculated effective growth coefficient from this growth process is  $k_{\text{eff}} = 3 \times 10^{-3}$  s/m. This number is actually very close to the value obtained from our measurements during

melting at the same temperature. The measured melting speed was  $1.67 \mu\text{m/s}$  for a corresponding overpressure of  $\Delta p = -11.3 \mu\text{bar}$  which yields  $k_{\text{eff}} = 2 \times 10^{-3} \text{ s/m}$ .

Thus the growth and melting velocities of rough surfaces were in our experiments about 2 times higher than the measured growth rate of the fastest facet (411). One can conclude that the processes determining the growth rates of the smooth surfaces exhibit intrinsic properties of the interface and are not limited by the thermal impedances of the bulk phases as in the case of rough surfaces.

To conclude, the growth rates of the faceted and rough surfaces were explored in  $^3\text{He}$  crystals. Both the growth and melting of the rough crystal surface yielded the effective growth coefficient of  $(2-3) \times 10^{-3} \text{ s/m}$  which is influenced by the thermal impedances of the bulk phases at a temperature of 0.55 mK [10]. The growth velocities of facets were much slower than those of rough surfaces and they revealed a significant anisotropy by differing more than an order of magnitude. The measurements exhibited a linear dependence of the facet velocity on the applied overpressure, which points to spiral growth in the regime of suppressed step mobility as the main growth mechanism.

The calculated step energies of the facets on  $^3\text{He}$  crystals feature a quartic dependence on the step height, suggesting that the steps experience elastic interactions. The step energy of the most "stable" (110) facet equals  $6.6 \times 10^{-10} \text{ erg/cm}$  and the corresponding step width  $w \sim 2a$  reflects relatively strong coupling of the interface to the crystal lattice.

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