

# Faceting and growth kinetics of $^3\text{He}$ crystals

Harry Alles<sup>1</sup>, Alexei Babkin<sup>2</sup>, Reyer Jochemsen<sup>3</sup>, Alexander Ya. Parshin<sup>4</sup>,  
Igor A. Todoshchenko, Viktor Tsepelin<sup>5</sup>

*Low Temperature Laboratory, Helsinki University of Technology, FIN-02015 HUT, Finland*

---

## Abstract

We review and discuss our recent results on faceting and growth kinetics of  $^3\text{He}$  crystals imaged with a Fabry-Pérot multiple-beam interferometer at our lowest temperature of 0.55 mK. More than ten different types of facets were identified on the growing crystals and the growth rates of the individual facets were measured. Some further possible experiments are also suggested.

*Key words:* Quantum crystals; helium-3; growth kinetics

---

## 1. Introduction

It has been shown by Landau that in the limit of zero temperature the surface tension of a crystal has a cusp for every rational (i.e., with integer Miller indices) orientation and that an ideal crystal is completely faceted at  $T = 0$  having an infinite number of facets, smooth flat faces, on its surface [1]. As temperature rises, these cusps are washed out in succession by thermal fluctuations and the corresponding surface parts enter the rough state (look rounded) at roughening transitions [2]. Above all roughening transition temperatures the equilibrium crystal shape is rounded and looks like a water droplet on a solid surface.

The experimental studies on the equilibrium crystal shape and on the roughening transitions are difficult with ordinary crystals which show only a few different types of facets and the growth dynamics of which can

be investigated only in a narrow temperature range close to their melting point.

The situation is different in helium crystals which can be studied in a wide temperature range along their melting curve down to  $T = 0$ . Therefore at the lowest temperatures they are surrounded by superfluid with good thermal conductivity which makes their dynamics very fast. The  $^4\text{He}$  crystals, for instance, relax to their equilibrium shape within milliseconds at  $T < 0.5$  K. In spite of cooling the  $^4\text{He}$  crystals down to 2 mK [3], no more but three types of facets have been seen in  $^4\text{He}$  [4].

In this paper the observations are described in which we have detected altogether eleven different types of facets on the slowly growing  $^3\text{He}$  crystals [5,6]. Before our experiments three types of facets had been seen in  $^3\text{He}$  [7], as in  $^4\text{He}$ . We have measured also the growth rates of the individual facets and calculated the step energies of facets by interpreting the data with the well-known spiral growth mechanism [8]. We discuss the consequences of our results and argue that  $^3\text{He}$  has much stronger coupling of the liquid/solid interface to the crystal lattice than  $^4\text{He}$ . And finally, we suggest some possible future experiments with  $^3\text{He}$  crystals.

---

<sup>1</sup> E-mail: harry@boojum.hut.fi

<sup>2</sup> Permanent address: Department of Physics and Astronomy, UNM, 800 Yale Boulevard NE, Albuquerque, NM 87131, USA

<sup>3</sup> Permanent address: Kamerlingh Onnes Laboratory, Leiden University, 2300RA Leiden, The Netherlands

<sup>4</sup> Permanent address: P.L. Kapitza Institute for Physical Problems, ul. Kosygina 2, 117334 Moscow, Russia

<sup>5</sup> Present address: Department of Physics, Stanford University, Stanford, CA 94305-4060, USA

## 2. Background

One of the most convenient methods to study the shape of helium crystals has been a visual observation through the windows of the conventional optical cryostat. In this way a systematic study of both the equilibrium and dynamic properties of  $^4\text{He}$  crystals has been performed [9]. However, the minimum temperature with that kind of setup is limited to about 20 mK and the dynamics of  $^3\text{He}$  crystals is at these temperatures still slow due to the large latent heat of crystallization and the poor thermal conductivity of the surrounding normal liquid. Only well below 1 mK, where the nuclear spins of solid  $^3\text{He}$  are in an (antiferromagnetically) ordered state and the surrounding liquid is superfluid, the mobility of the superfluid/solid interface of  $^3\text{He}$  becomes comparable to that of  $^4\text{He}$  at much higher temperatures.

The optical observations at submillikelvin temperatures became feasible in the beginning of 1990ies when a new technical approach was put forward by two research groups in Leiden [10] and in Helsinki [11]. The conventional optical cryostat was replaced with a setup where the imaging system was built inside the cryostat so that there was no direct access of the infrared radiation from the room temperature to the sample.

In this novel type of optical cryostat two new types of facets, (100) and (211), were detected on the body-centered cubic (*bcc*)  $^3\text{He}$  crystals grown from the superfluid phase [7], additionally to the (110) facets seen at higher temperatures [12]. The growth rates of individual facets were not measured in these experiments. The equilibrium shape of  $^3\text{He}$  crystals has been observed only for rounded crystals in the vicinity of the melting curve minimum where the latent heat is small [13].

## 3. Imaging the crystals using the phase-shift technique

For our experiments we have modified the setup used by the Helsinki group to study the  $^4\text{He}$  crystals with a two-beam Fizeau interferometer [14]. In order to image the  $^3\text{He}$  crystals a Fabry-Pérot multiple-beam interferometer was designed and built inside the nuclear demagnetization cryostat [15].

The interferometer consists of two semitransparent mirrors above and below the cylindrical ( $\varnothing = 16$  mm) optical cell which is sealed off with antireflection-coated windows. The He-Ne laser beam ( $\varnothing \approx 8$  mm) enters the interferometer through the upper mirror. The light undergoes multiple reflections between the mirrors and the interference pattern (see Fig. 2) is focused to a slow-scan CCD-sensor inside the 4-K vacuum can of the cryostat. The phase shift techniques

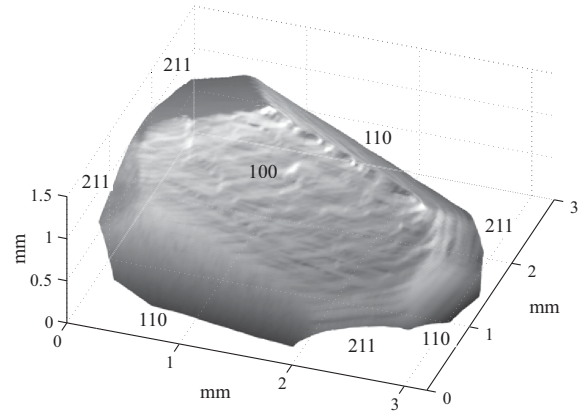


Fig. 1. Reconstructed surface profile of a  $^3\text{He}$  crystal at 0.55 mK obtained using the phase shift technique. The identified facets are labeled with Miller indices.

can be applied in our setup thanks to the movable upper mirror glued to a piezo crystal to which a high voltage can be applied. For more details of our experimental setup, we refer to Ref.[16].

The phase shift techniques allow us to measure (by taking several interferograms with a known phase shift in between) the phase map of the imaged crystal from which the crystal thickness plot can be calculated [15,16]. This thickness plot corresponds to the real crystal profile when the lower part of the crystal (against the optical window) is a single flat surface. As an example, in Fig. 1 the reconstructed surface profile of a  $^3\text{He}$  crystal, showing three different types of facets, is depicted. In dynamic situations, however, the phase shift-techniques cannot be used and in order to analyze single interferograms we have used the intensity based analysis methods (see next Section).

## 4. Observations of higher order facets and measurements on the growth anisotropy of $^3\text{He}$ crystals

The higher order facets were observed in our experiments when growing slowly big (several millimeters in size) crystals starting from the rounded shape obtained by melting. Figure 2 shows two interferograms of a growing  $^3\text{He}$  crystal at 0.55 mK. The sets of equidistant and parallel fringes on the interferogram (except the background pattern) correspond to facets on the crystal surface; the adjacent fringes indicate a change of  $190 \mu\text{m}$  in the crystal thickness. The normal vectors of the facets were calculated from the interferograms using the intensity based analysis methods [6]. The facet types were identified by comparing the measured angles between the facets with the theoretically

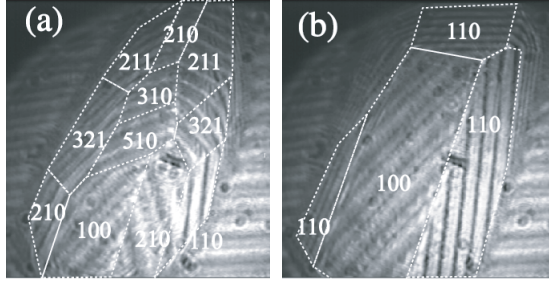


Fig. 2. Interferograms of a growing  $^3\text{He}$  crystal (a) from the middle and (b) from the end of the growth sequence. The dashed white lines outline the identified facets marked with Miller indices.

possible ones.

In Figure 2a, in the middle of one of a typical growth sequence, seven different types of facets can be counted. In a course of growth the facets continuously changed in size and appeared/disappeared until finally only a few large (lower order) facets remained as in Fig. 2b. In our experiments we detected in total eleven different types of facets which are shown in Fig. 3 on an elementary patch of the crystal habit.

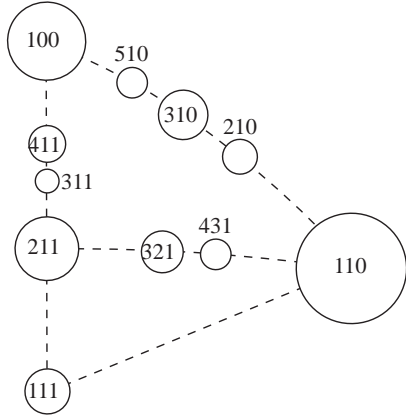


Fig. 3. Diagram with the observed facets on  $^3\text{He}$  crystals. The areas of the circles are proportional to the expected roughening transition temperatures.

Additionally to the identification of new facets we were able to measure the growth velocities  $v$  of the individual facets as functions of overpressure  $\delta p$  and obtained a linear dependence as seen from Fig. 4. We interpreted our data with the spiral growth in the regime of so called limited step mobility [8]. In that regime

$$v = \frac{v_c d^2}{2\pi\beta} \frac{(\rho_s - \rho_l)}{\rho_l} \delta p, \quad (1)$$

where  $\rho_s$  and  $\rho_l$  are the densities of the solid and liquid,  $d$  is the elementary step height on a facet (the interplanar distance of the corresponding orientation) and  $v_c$  is the critical velocity. Using the measured growth

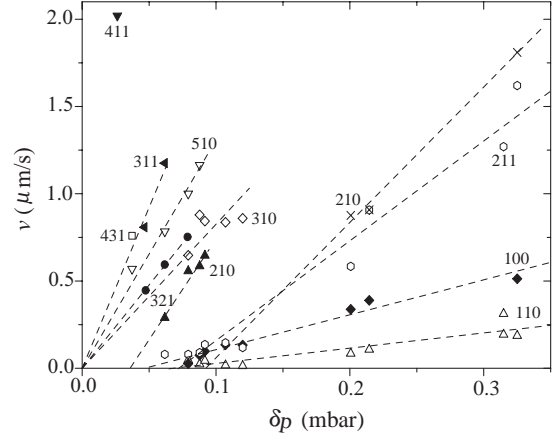


Fig. 4. Growth anisotropy of  $^3\text{He}$  crystals at  $T = 0.55$  mK. The dashed lines are the linear fits to the data.

rates and Eq. (1) the step energies  $\beta$  can be calculated for different facets because the lowest critical velocities in  $^3\text{He}$  are the magnon velocity in the solid and the pair-breaking velocity in the liquid which both are about 7 cm/s below 1 mK [17,18]. The obtained results are plotted in Fig. 5. We found that the step energies of facets vary like the fourth power of the corresponding step height which refers to the elastic interaction between steps [19].

## 5. Discussion

The step energy is proportional to the equilibrium size of the facet [1]. Thus according to our data the fast-growing higher order facets have very small step ener-

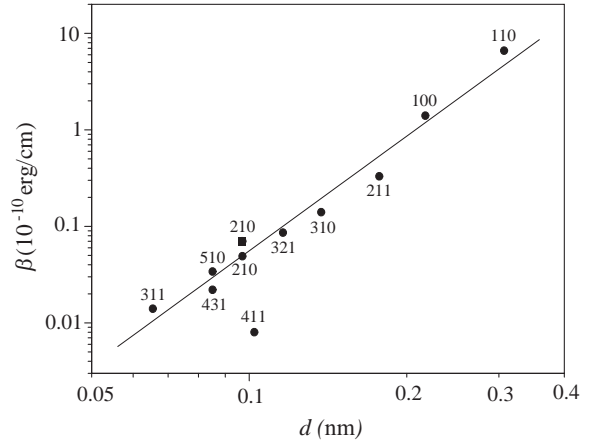


Fig. 5. Calculated step energies of different facets plotted versus the corresponding step heights. The linear fit (except for the (411) facet) has a slope of  $3.95 \pm 0.25$ .

gies and very small equilibrium sizes. This might be actually the reason why no higher order facets has been detected on  $^4\text{He}$  crystals which have extremely short relaxation time at low temperatures. In  $^3\text{He}$  we have been able to see many different types of facets because we observed the facets during growth, far from equilibrium. On the other hand, the step energy of the (110) facet,  $\beta_{110} = 6.6 \cdot 10^{-10}$  erg/cm, having the largest interplanar distance in the *bcc* lattice, yields the equilibrium size for that type of facet of about one third of the characteristic size of the crystal [6], which should be easily measurable.

The value of the step energy of the (110) facet is of the same order as for the basal (1000) facet on the hexagonal close packed  $^4\text{He}$  crystals. While the measured surface tension of  $^3\text{He}$  crystals (0.06 erg/cm<sup>2</sup> [13]) is about four times smaller than in  $^4\text{He}$  that points to much stronger coupling of the crystal lattice to the liquid/solid interface in the case of  $^3\text{He}$ . Also the fact that we were able to see so many different facets supports the conclusion that this coupling is strong in  $^3\text{He}$ .

## 6. Future experiments

(1) The existence of a multitude of different facets could make  $^3\text{He}$  very useful system to check the general theories related to the roughening transition. For instance, it would be very interesting to measure the roughening transition temperatures of the observed facets. These kind of measurements have been carried out for the (110) facets and the corresponding roughening transition temperature (or the lower limit of it) was found to be about 100 mK [13]. Additionally to that we could measure the surface stiffness of the rounded  $^3\text{He}$  crystals by our interferometric technique. One has to keep in mind, however, that in  $^3\text{He}$  in this temperature range the situation is very similar to ordinary crystals, i.e., the dynamics of crystals is very slow and the experiments are thus difficult.

(2) The growth dynamics of  $^3\text{He}$  crystals could be studied in the neighborhood of the antiferromagnetic transition of solid  $^3\text{He}$  at 0.93 mK where the disordered nuclear spins of solid  $^3\text{He}$  transform into a tetragonal structure with ferromagnetically polarized (100) planes [20]. It would be natural to expect a polarization induced change in facet mobilities at the transition.

(3) We believe that we could measure the equilibrium sizes of the lower order facets on  $^3\text{He}$  crystals. The measurements on the equilibrium shape of a faceted  $^3\text{He}$  crystal would therefore be easier at temperatures well below 0.5 mK because the mobility of the su-

perfluid/solid interface of  $^3\text{He}$  increases considerably towards lower temperatures.

## 7. Conclusions

In our interferometric experiments with  $^3\text{He}$  crystals we have identified eleven different types of facets and measured the growth anisotropy of the crystals. Our interpretation of the obtained results yields conclusion that in spite of higher zero-point oscillations  $^3\text{He}$  has much stronger coupling of the liquid/solid interface to the crystal lattice than  $^4\text{He}$ .

## Acknowledgements

We would like to thank J.P.H. Härme and G. Tvalashvili and for their contribution in the early stage of the project. This work was supported by the ULTI II (ERB-FMGE-CT-98-0122) and ULTI III (HPRI-1999-CT-00050) grants of the European Union and the Academy of Finland and by INTAS Grant 96-610.

## References

- [1] L.D. Landau, *Collected Papers* (Pergamon Press, Oxford, 1971).
- [2] W.K. Burton, N. Cabrera, and F.C. Frank, *Philos. Trans. R. Soc.* **243** (1951) 299.
- [3] J. P. Ruutu *et al.*, *J. Low Temp. Phys.* **112**, 117 (1998).
- [4] P.E. Wolf *et al.*, *Phys. Rev. Lett.* **51** (1983) 1366.
- [5] V. Tsepelin *et al.*, *Phys. Rev. Lett.* **86** (2001) 1042.
- [6] H. Alles *et al.*, *J. Low Temp. Phys.* **124** (2001) 189.
- [7] R. Wagner *et al.*, *Phys. Rev. Lett.* **76** (1996) 263.
- [8] V. Tsepelin *et al.*, *Phys. Rev. Lett.* **88** (2002) 045302.
- [9] S. Balibar and P. Nozières, *Solid State Com.* **92** (1994) 19 and references therein.
- [10] R. Wagner *et al.*, *J. Low Temp. Phys.* **95** (1994) 715.
- [11] A.J. Manninen *et al.*, *Phys. Rev. Lett.* **69** (1992) 2392; H. Alles *et al.*, *Rev. Sci. Instrum.* **65** (1994) 1784.
- [12] E. Rolley *et al.*, *Europhys. Lett.* **2** (1986) 247.
- [13] E. Rolley *et al.*, *Europhys. Lett.* **8** (1989) 523.
- [14] P. Hakonen *et al.*, *Czech. J. Phys.* **46** (1996) 2965.
- [15] J.P.H. Härme *et al.*, *Physica B* **284-288** (2000) 349.
- [16] V. Tsepelin *et al.*, *J. Low Temp. Phys.* **121** (2000) 695.
- [17] D.D. Osheroff and C.Yu, *Phys. Rev. Lett.* **77** (1980) 458.
- [18] Y.P. Feng *et al.*, *J. Low Temp. Phys.* **90** (1993) 475.
- [19] V.I. Marchenko and A.Ya. Parshin, *JETP* **52** (1980) 129.
- [20] Y. Sasaki *et al.*, *Phys. Rev. B* **44** (1991) 7362.