

Dynamics and morphology of superfluid bubbles in ^4He quantum crystals

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Superfluid bubbles in a ^4He quantum crystal, which we refer to as quantum negative crystals, were investigated in bcc and hcp phases by visualizing their forms and motions at various temperatures. They were nearly spherical in bcc phase and faceted on the c -plane in their upper part in hcp phase. They steadily rose in the crystal due to gravity. Their direction was vertical in bcc phase and obliquely parallel to the c -facet in hcp phase. We also observed a slowing down of negative crystal in hcp phase caused by the appearance of the a -facet when temperature was lowered. Dynamics and morphology became successively anisotropic with cooling. The driving force by gravity for this motion was derived in the spherical case. Growth coefficient obtained by this model agreed well with the reported values.

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I. INTRODUCTION

^4He quantum crystal in a superfluid is an ideal system to investigate fundamental properties of the crystal-liquid interface due to its very quick response to a driving force [1,2]. Superfluid can transport mass and heat, which are necessary for the crystallization to proceed, very quickly. Mobility of the interface is limited by the crystallization kinetics at the interface, but is not by the bulk transport. Mobility is determined by the collision between the interface and thermal rotons and phonons [3–5]. Densities of these quasiparticles decrease at low temperatures and the mobility becomes anomalously large compared with the ordinary materials. Relaxation time of the interface is short enough to investigate the static and dynamical properties very precisely. It has contributed greatly to the understanding of physics in crystal growth. To date three roughening transitions have been observed in hcp phase for c -facet, a -facet, and s -facet whose transition temperatures are $T_{R1}=1.3$ K, $T_{R2}=0.9$ K, and $T_{R3}=0.36$ K, respectively [6–9].

A liquid or gas bubble in a crystal is called a negative crystal when the form of the bubble is anisotropic and surrounded by faceted surfaces. Equilibrium form of the negative crystal is expected to be the same as that of real crystal in order to meet the conditions for minimizing the surface free energy. A negative crystal can exist inside the crystal without touching walls or substrates, while a real crystal usually touches the wall because of gravity and is deformed by the wall. Achieving equilibrium in a small negative crystal is relatively easy because the process proceeds in the small space. Studying the form of the negative crystal is expected to be useful in order to see the equilibrium thermodynamic phenomena like equilibrium form, roughening transition and so on. Nelson *et al.* discussed theoretically the equilibrium forms and size of negative crystals [10]. Equilibrium forms were observed in negative crystals in diphenyl crystals by Pavlovskaya and Nenow [11]. Growth forms and surface kinetics were investigated in negative crystals in ice by Knight

and Knight [12] and by Furukawa and Kohata [13].

Steady motion of the negative crystal in the crystal is quite different from that of the crystal in the fluid due to the conservation of mass in the negative crystal: one side of the negative crystal is melted and the other is crystallized in order to move the center of the mass of the negative crystal. Negative crystal motion is inevitably accompanied by melting and crystallization. Direction of the flow of the fluid in the negative crystal is opposite to the direction of the negative crystal motion if the density of the fluid is less than that of the crystal. It is interesting to see how the dynamics and morphology are related to the surface properties of the real crystal such as roughening transition, anisotropy in mobility and so on. We were able to investigate the dynamics and morphology of ^4He negative crystal in this manner of steady motion, although the motion of the negative crystal in ordinary materials would be too slow to make a systematic study. A negative crystal in quantum solid ^4He , which was filled with superfluid, moved reasonably fast so that we could investigate its dynamics in a reasonable experimental time due to the large mobility of the interface at low temperatures. It is appropriate to call it a quantum negative crystal. Several groups have reported on inclusions in ^4He crystal. Keshishev *et al.* observed faceted bubbles in ^4He crystal which were created by applying high voltage to a capacitor [6]. Polturak *et al.* observed boiling of bubbles in solid ^3He - ^4He mixtures [14]. Okuda *et al.* observed dynamic motion of faceted bubbles in ^4He crystal which were created by acoustic waves [15].

A ^4He negative crystal, when it was created in the lower part of a ^4He crystal, rose in the crystal to the bulk crystal-superfluid interface because the negative crystal was lighter than the crystal. Motion and form of the negative crystal was recorded by a video camera by changing temperature, crystal orientation, and the size of the negative crystal. In bcc phase, which is more isotropic than hcp phase, the negative crystal was nearly spherical and rose vertically. No anisotropy in dynamics or morphology was observed. In hcp phase below the roughening transition of the c -facet, the form of negative crystal was drastically changed: a large c -facet appeared in the upper half and the lower half was rounded. The upper

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part was melted and the lower part was crystallized for the negative crystal to rise in the crystal. It is a well-known rule for a positive crystal that facets expand on a crystallizing surface and shrink on a melting one. The rule is reversed for a negative crystal and thus the strange form of the negative crystal resulted from the melting and crystallization. The negative crystal in hcp crystal rose not vertically but obliquely parallel to the c -facet because of the small mobility of the c -facet. At lower temperatures below the roughening transition of the a -facet, the speed of the negative crystal motion was decreased. This is because the a -facet appeared laterally in the direction of the negative crystal motion. We clearly demonstrated that dynamics and morphology of the negative crystal became successively anisotropic as the temperature lowered.

II. EXPERIMENTS AND RESULTS

We performed experiments in a ^3He refrigerator for bcc crystals at high temperatures and in a dilution refrigerator for hcp crystals at low temperatures. Both refrigerators had windows so that the crystals in the experimental cells could be observed visually from room temperature. Superfluid ^4He was supplied to the cell through a capillary and pressurized to melting pressure. A large crystal was grown from the bottom of the cell and occupied the lower space. The crystal was grown up to fill the most of view through windows, but enough superfluid was left in the upper space of the cell to ensure that the system was at melting pressure. In the case of the cell in the ^3He refrigerator, negative crystals spontaneously and continuously appeared on the bottom of the cell, probably because of a slight temperature gradient in the cell. The top of the cell was anchored to the refrigerator and was colder than the bottom. This did not happen in the cell in the dilution refrigerator whose bottom was anchored to the mixing chamber and was coldest. A negative crystal never appeared spontaneously in this cell. An acoustic wave pulse was used for this cell to nucleate a negative crystal in the crystal. This negative crystal appeared on the acoustic transducer. The size was roughly adjustable by the power and period of the acoustic wave. Experimental setups were the same as reported in our previous publications [16,17]. Details of the nucleation of negative crystal by this method will be published elsewhere [18].

Once a negative crystal appeared on the bottom of the cell or on the transducer, it began to rise steadily in the crystal to the crystal-liquid interface. This is because the density of the liquid or the negative crystal is less than that of the crystal. Thus the driving force of this motion is gravity. When the negative crystal reached the crystal-liquid interface it coalesced with the interface and disappeared; Fig. 1 shows from the birth to the disappearance of a negative crystal. Diameter of the negative crystal was 2.8 mm and $T=1.66$ K. We analyzed this series of images for many negative crystals to measure their rising speed with changing temperature, crystal orientation and size.

An enlargement of a spherical negative crystal in bcc crystal is shown in the inset of Fig. 2. The direction of motion of a negative crystal in bcc phase was always vertical as

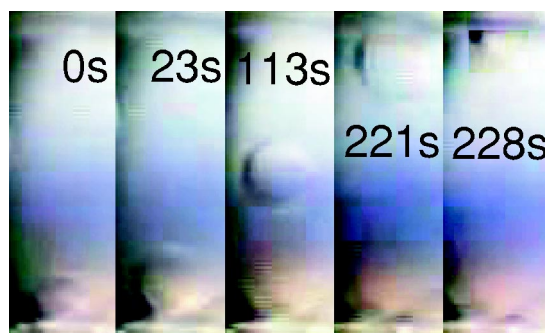


FIG. 1. (Color online) Successive images of a spherical negative crystal in bcc phase from its birth to disappearance. It appeared on the bottom of the cell and rose vertically in the crystal due to gravity, finally reaching the crystal-liquid interface and disappearing. The diameter of the negative crystal was 2.8 mm and $T=1.66$ K.

indicated by an arrow in the inset. The morphology and the dynamics were quite isotropic and were not influenced by the orientation of the host crystal. It looked as if a bubble rose in a very viscous fluid due to the buoyancy. Figure 2 indicates the temperature dependence of the rising speed v of negative crystals in bcc phase whose diameters were in the range of 2.7 ± 0.3 mm. It was important to choose the proper size of the negative crystals to compare v because the larger ones moved faster than the smaller. We could not control their size in this cell for bcc crystals and thus could show v of the negative crystals only in this range. Although the scatter is a little large due to their distribution of the size, it can be seen that negative crystals moved faster at lower temperatures. As mentioned above, the motion of the negative crystal is inevitably accompanied by crystallization and melting at the surface, and this temperature dependence is closely related to the temperature dependence of the crystal growth coefficient $K(T)$. Comparison with the reported values of $K(T)$ will be made in the discussion section assuming a simple driving force of gravity for the rise of negative crystal.

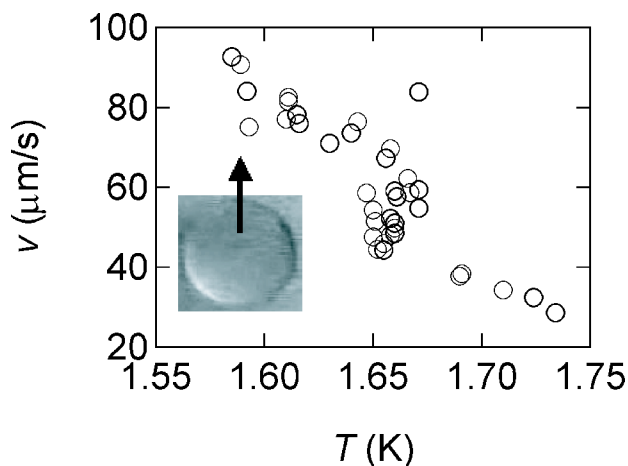


FIG. 2. Temperature dependence of the velocity of negative crystals in bcc phase. Only the data of negative crystals with a diameter in the range of 2.7 ± 0.3 mm were plotted. Inset is an expanded image of a spherical negative crystal and arrow indicates the direction of its motion.

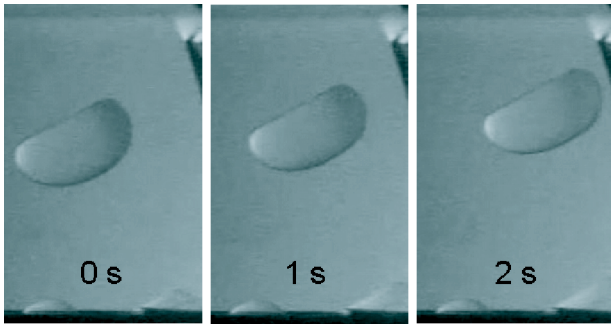


FIG. 3. Successive images of a negative crystal in hcp phase at $T=0.96$ K. The width was 3.6 mm in the widest direction. A large c -facet appeared in the upper part. It moved obliquely parallel to the c -facet. Form and motion became highly anisotropic in hcp phase.

Form and motion of negative crystals were drastically changed at low temperatures in hcp crystal as shown in Fig. 3. These images were taken at $T=0.96$ K. A large c -facet, the roughening temperature of which is known to be $T_{R1}=1.3$ K, appeared in the upper part of the negative crystal and a rounded rough surface in the lower part. It steadily rose in the crystal due to gravity, not vertically but obliquely parallel to the c -facet. Curvature of the rough surface was a little larger on the rear side than the advancing side. Morphology and dynamics became highly anisotropic and were influenced by the static (surface stiffness) and the dynamical (anisotropy in $K(T)$) properties of the host crystal. Because we could control the size of the negative crystals in hcp phase by adjusting the power and the period of the acoustic wave, we were able to measure the size dependence of the obliquely rising speed v . The size dependence of v of negative crystals at $T=0.92$ K is shown in Fig. 4. The velocity v was measured along the crystal motion which was parallel to the c -facet. In order to represent their size we took the effective height of the negative crystal $2R \sin \theta$ as illustrated in the inset of Fig. 4. The larger these crystals were, the faster they moved. Linear dependence of v on the effective height can be seen clearly. Figure 5 shows temperature dependence of v for negative crystals, which had almost the same size with the effective height in the range of 3.4 ± 0.3 mm. This is a different crystal from that shown in Fig. 4. The velocity v

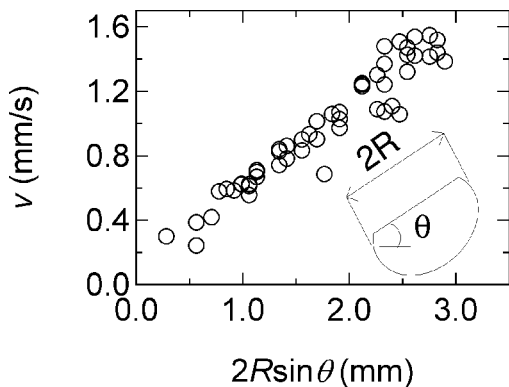


FIG. 4. Size dependence of the velocity of negative crystals in hcp phase at $T=0.92$ K. Effective height $2R \sin \theta$ was determined as in the inset. It increased linearly to the effective height.

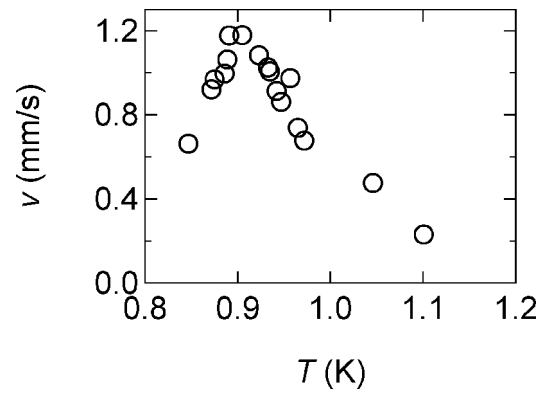


FIG. 5. Temperature dependence of velocity of negative crystals whose diameters were in the range of 3.4 ± 0.3 mm. Decrease of v below 0.9 K is due to the appearance of the a -facet.

became larger as T lowered and started to decrease below 0.90 K, which corresponds to the roughening transition temperature of the a -facet, $T_{R2}=0.9$ K. We believe this decrease at low temperatures was caused by the appearance of the a -facets on the lateral position of the negative crystals. Unfortunately, no clear images of a -facets were obtained for the negative crystals shown in Fig. 5 presumably due to a bad camera angle. Also, because the a -facet is weaker or smaller than the c -facet it was difficult to see them clearly. However, we succeeded in obtaining better images of the a -facet for another negative crystal with a different orientation; in this case the camera was slightly looking down on the c -facet of this negative crystal and images are shown in Fig. 6. Above T_{R2} the surface in the direction of the negative crystal motion was rounded as in Fig. 6(a). Below T_{R2} an a -facet on the opposite side of the negative crystal can be seen through the negative crystal as a dark image in the dotted circle of Fig. 6(b).

Note that we observed the motion of negative crystals only from one direction and ignored their approaching or retreating motion. We did our best to choose crystals in which the negative crystal motion was almost perpendicular to our view. There exist systematic errors in v of less than $\pm 20\%$ when we compare data of crystals with different orientation. This was not a problem in bcc phase because negative crystal always rose vertically.

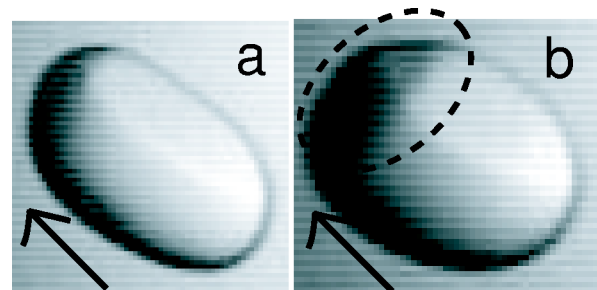


FIG. 6. Expanded images of negative crystals at $T=0.92$ K (a) and $T=0.87$ K (b), which are above and below the roughening transition of the a -facet. Arrows indicate their direction of movement. We can see the a -facet on the opposite side the negative crystal looking through it as a dark image in the dotted circle in (b), which does not exist in (a).

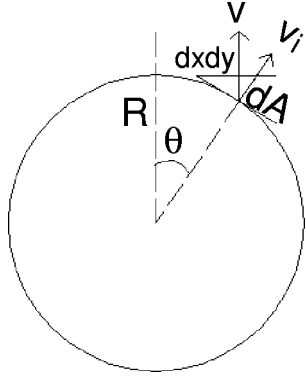


FIG. 7. Steady motion of a spherical negative crystal with a radius of R . It rises with a vertical velocity of v and a normal velocity of the interface v_i . Small area on the sphere dA and its projection $dx dy$ are defined as in the figure.

III. DISCUSSION

Let us consider the steady motion of a spherical negative crystal or a liquid bubble with a radius of R in a crystal driven by gravity. The steady motion of the sphere with rising speed of v is realized by the uniform downward flow of the fluid in the negative crystal as shown below [19]. We assume that the motion is slow enough to neglect the kinetic energy of the fluid. A small area on the sphere dA is related to the projection on the x - y plane $dx dy$ as

$$dA \cos \theta = dx dy \quad (1)$$

if θ is the polar angle as in Fig. 7. Normal velocity of the interface v_i can be written as

$$v_i dA = v \cos \theta dA = v dx dy. \quad (2)$$

Chemical potential difference between dA and the surface beneath is given as

$$\Delta\mu = 2\Delta\rho ghR \cos \theta, \quad (3)$$

where $\Delta\rho = \rho_c - \rho_l$ and g are density difference between crystal and liquid and the gravity acceleration. This driving force moves the interface as

$$v_i = K(T)\Delta\mu. \quad (4)$$

Comparing Eqs. (1), (3), and (4),

$$\begin{aligned} v_i dA &= K(T)\Delta\mu dA = 2K(T)\Delta\rho gR \cos \theta dA \\ &= 2K(T)\Delta\rho gR dx dy. \end{aligned} \quad (5)$$

From Eqs. (2) and (5)

$$v = 2K(T)\Delta\rho gR. \quad (6)$$

Equation (6) is independent of θ and thus a steady motion of the sphere is realized in this model. Uniform superflow v_l in the sphere is obtained by considering the conservation of mass at the interface as

$$v_l = -\frac{\Delta\rho}{\rho_l}v. \quad (7)$$

From Eq. (6) we can see that negative crystal with large R rises faster in the crystal. This is consistent with our obser-

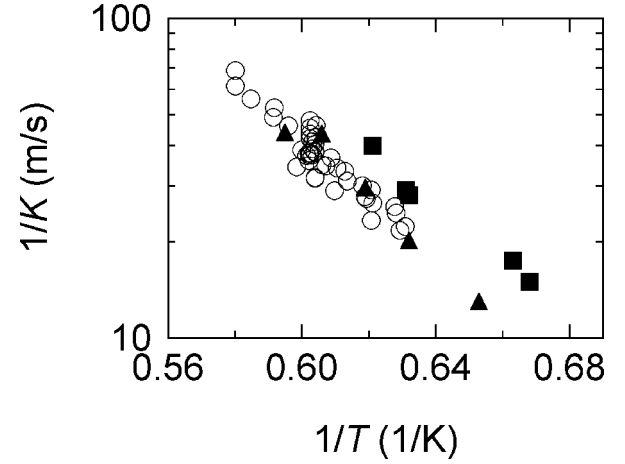


FIG. 8. Temperature dependence of the crystal growth resistance $1/K$ in bcc phase obtained from Eq. (6). Reported values by Bodensohn *et al.* and Tsymbalenko are also plotted as squares and triangles, respectively. They reasonably agree with each other.

vation in bcc phase that larger negative crystals did rise faster. However, linear dependence of v on R could not be checked with good accuracy because we could obtain the negative crystals only in a limited range of sizes. As mentioned, the negative crystals in bcc phase appeared spontaneously on the bottom of the cell and their size was not controllable. In hcp phase, however, we were able to control the size of these negative crystals over a wider range. Linear dependence on the effective height $2R \sin \theta$ was clearly obtained as in Fig. 4. Although it is not obvious that motions of negative crystals in hcp phase with the oblique form can be compared with the simple sphere model, linear dependence is expected to be observable due to the same driving force, which is gravity, if their effective height is properly estimated. It is not too much to say that Eq. (6) is partly confirmed by Fig. 4. An experiment in bcc phase over a wider range of R is needed to confirm Eq. (6) more strictly.

Using Eq. (6) it is possible to convert the temperature dependence of v in Fig. 2 to $K(T)$ and to compare them with $K(T)$ in the literature. Figure 8 is obtained by this conversion using the radius of each negative crystal. Scatter in the data originating from the size distribution was slightly improved by this conversion. Reported values by Bodensohn *et al.* [20] and Tsymbalenko [21] are also plotted as squares and triangles, respectively. Agreement with the reported values is reasonably good and this is further support of Eq. (6). In hcp phase Eq. (6) is too simple to convert v in Fig. 5 to $K(T)$ and to make a serious comparison with the literature values. In order to do so we need a precise numerical coefficient between v and $K(T)$ taking into account the anisotropic properties of the interface, although simple analytical calculation cannot be expected.

It is not difficult to understand intuitively the strange form of the negative crystals in the hcp crystal. Mobility of the c -facet is much smaller than that of the rough surface. When a negative crystal rises, the slow surface is caught up with by the fast surface in the upper part of the negative crystal. This results in widening of the c -facet. In the lower part of the negative crystal the rough surface gets ahead of the facet and

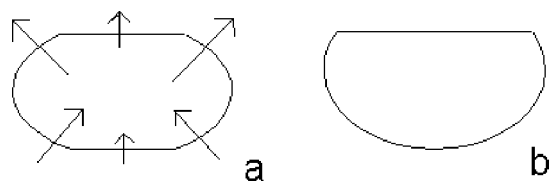


FIG. 9. Intuitive explanation for the form of negative crystal in motion in hcp phase. Length of the arrows in (a) indicate the velocity of the interfaces. Rough surfaces move faster than c -facets. This results in widening of the c -facet in the upper part of the negative crystal and the shrinkage in the lower part as shown in (b).

the facet shrinks. This situation is illustrated in Fig. 9. Generally speaking the widening of facets is a characteristic of the growth form of crystal and their shrinkage is characteristic of the melting form. The motion of negative crystal is inevitably accompanied by the melting in the moving direction and by the crystallization in the opposite direction. It is reasonable that moving negative crystal has a hybrid character of growth and melting forms. What happens to the form and motion of a negative crystal at around roughening transition temperature of the c -facet, T_{R1} ? This is an intriguing problem which we have not yet investigated.

Dynamics of disappearance of the negative crystal at the crystal-liquid interface is another feature we have not explored in detail. Recently Maris predicted that coalescence of two liquid drops, if their interface motion is limited not by viscosity of the fluid but by its interface mobility, is different from that of normal viscous fluid [22]. The neck radius that forms between two liquid drops is proportional to $t^{1/3}$, where time t is measured from the moment of impact. Ishiguro *et al.* have shown quantitative agreement with the prediction in coalescence of ^3He crystals at the minimum of the melting curve [23]. Negative crystal, whose mobility is determined by $K(T)$ as we demonstrated in this paper, can be another

example described by Maris' theory. We have expertise of getting high-speed images to investigate crystal growth dynamics in ^4He [16,17] and will be able to apply the technique to study the coalescence of two negative crystals in ^4He .

IV. SUMMARY

Dynamics and morphology of negative crystals in quantum solid ^4He , which were referred to as quantum negative crystals, were investigated by directly visualizing them at low temperatures both in bcc and hcp phases. They moved up in the crystal due to gravity. In bcc phase negative crystals were spherical and rose vertically. In hcp phase a large c -facet appeared in the upper part of the negative crystals, and they moved not vertically but along this c -facet. Velocity of the negative crystal increased with cooling in both phases in high temperature regions but decreased below 0.9 K due to the roughening transition of the a -facet in hcp phase. Dynamics and morphology of the negative crystals became successively anisotropic by cooling. Driving force of the motion was derived in the case of the spherical negative crystal. Temperature and size dependence of the motion was consistent with this model and the mobility of the interface in the literature was well reproduced by this model in bcc phase.

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