Falling $^4$He crystals in superfluid

Ryuji Nomura, Taichi Yoshida, Akira Tachiki and Yuichi Okuda
Department of Physics, Tokyo Institute of Technology, 2-12-1, O-okayama, Meguro-ku, Tokyo 152-8551, Japan
E-mail: nomura.r.aa@m.titech.ac.jp

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Abstract
The crystal shapes of $^4$He falling in superfluid were investigated using a high speed video camera. As they fell, the upper surface of the crystal became rough and the lower surface became faceted as a result of being subjected to an upward superflow which induced melting of the upper surface and crystallization of the lower surface. When it landed on the bottom, a pulse-like wave was observed to travel around the surface from the impact point before it transformed itself quickly to adjust to a new boundary condition. The behavior of a crystal when it passed by a small needle during its fall was also observed. The rim of the facet was pulled by the needle and protruded. The origin of this extended deformation is not understood at present, but the superflow around the crystal was possibly the origin of the interaction between the crystal and the needle.

Online supplementary data available from stacks.iop.org/njp/16/113022/mmedia
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1. Introduction

Hydrodynamics and interfacial instabilities of Bose-condensed systems have drawn increasing interest recently for use in examining universal features of non-equilibrium fluid dynamics from the viewpoints of the established elementary processes of order-parameter fields [1–5]. In
contrast, first order phase transition of classical matter under a mass or heat flow is very difficult to understand and is actively discussed in statistical physics to look for general descriptions of non-equilibrium systems \[6–9\]. $^4$He quantum crystals growing from superfluids could possibly be a system to bridge the two circumstances. Since the crystal growth rate of $^4$He is very high at low temperatures \[10, 11\], crystal shapes are thought to be sensitive to the superflow surrounding the crystals, thus opening a new possibility that the effect of the mass flow on the first order phase transition can be visualized by the change in crystal shape.

A few experiments have been performed to investigate the effect of superflow \[12, 13\], however, it is not easy to induce a uniform and stable superflow experimentally. It is intriguing to see how the crystal shape of $^4$He is changed by crystallization and melting when subjected to a superflow, responding almost instantaneously to the surrounding superflow due to the high crystal growth rate. This is in contrast to the growth of classical crystals such as snow crystals. The shape of snow crystals changes slowly, and thus their shapes on the ground are closely connected to their history or the condition in which these crystals were formed in the sky \[14\]. The shape of $^4$He crystals, however, is connected to the simultaneous superflow and changes every moment adjusting itself to the external conditions. Here, we report on the crystal shapes of $^4$He falling in superfluid as a result of gravity. The crystals were subjected to a rather uniform superflow from beneath and their shape changed quickly and drastically.

2. Experimental procedure

Experiments were performed in high pressure sample cells cooled by a $^3$He–$^4$He dilution refrigerator with an optical access. Behavior of $^4$He crystals inside the sample cell was observable from room temperature; crystals were illuminated through the back windows of the refrigerator and observed from the front side by a high-speed camera. An ultrasound transducer was installed in the upper part of the cell and used to nucleate a $^4$He seed crystal on its surface. Superfluid $^4$He was supplied through a capillary to pressurize the cell a few mbars above the crystallization pressure. An ultrasound pulse was emitted from the transducer in the metastable superfluid in order to induce nucleation of the crystal. Right after the pulse a $^4$He crystal was nucleated on the transducer surface. The experimental setup and the nucleation procedure were described in previous publications \[15, 16\].

Superfluid $^4$He was continuously supplied to the cell to grow the $^4$He seed crystal to a proper size on the transducer; once achieved, we stopped the supply and waited a couple of minutes. The crystal relaxed to an equilibrium shape in which the corners between the facets in the lower part were rounded as shown in figure 1(a). The temperature was 0.3 K. The crystal shape was a hexagonal prism whose upper part was cut by the transducer obliquely. The top and bottom surfaces of the prism were c-facets and the side surfaces were a-facets. It was too large to remain attached to the transducer and eventually fell in the superfluid as in figures 1(b–o). The distance between the transducer and the bottom where the crystal finally landed in figures 1(p–r) was 10 mm.

3. Results and discussion

As the crystal fell in the superfluid, its rounded corners became sharper in the lower surface; the facets became larger and the rough surface between them shrank, as is clearly seen in figures 1(h–o). At the same time, its upper part which was initially rounded became more
horizontal and nearly flat with some undulations; this upper surface was actually a rough surface. The shape of the crystal changed during its fall. A video clip showing the entire process in figure 1 is provided as supplementary data (available from stacks.iop.org/njp/16/113022/mmedia). This falling motion was quite different from that observed at the much higher temperature of 0.9 K in [17], in which a much thinner crystal rotated as it fell.

In a frame moving with the $^4$He crystal, it was subjected to the upward superflow. This superflow is the origin of the drastic change in shape of the falling crystal. The lower surface of $^4$He crystal faced the incoming superflow which induced the crystal growth, while the upper surface saw off the outgoing superflow which induced the melting. The enlargement of the facet and that of the rough surface are known typical behaviors of the growth and melting shape of crystals, respectively, due to very large anisotropy in their growth rate [18, 19]. Generally, rough surfaces have a high growth rate and facets have a low growth rate. A peculiar point in the falling crystal is that both growth shape and melting shape appeared at the same time in the upper and lower parts of the single crystal due to the unidirectional upward superflow.

Once the crystal hit the bottom its shape quickly changed in order to adjust to the new boundary condition as in figures 1(p–r); usually the contact angle between a $^4$He crystal in superfluid and a wall is 135° [10, 11]. In figure 2, enlarged images of the crystal around the

Figure 1. Falling of a $^4$He crystal in superfluid at 0.3 K. Times are indicated in each frame. The lower surface was covered by the facets due to the incoming superflow and the upper surface was rough due to the outgoing superflow during the fall. The crystal quickly deformed to adjust to a new boundary condition set by the bottom after landing.
impact are shown in shorter periods of 1 ms. A pulse-like surface wave propagated on the crystal as indicated by the arrows. Surface waves on 4He crystals have been called crystallization waves which occur because of the high crystallization rate [20, 21]. The pulse-like crystallization wave was generated by the impact with the wall and propagated from the impact point along the surface; it had a velocity of about 0.7 m s\(^{-1}\).

Impacts between objects, which are elastic or dissipative, are important in diverse fields such as geosciences and the physics of powder. The impact shown in figure 2 is another intriguing limit in which the effects of the elasticity and dissipation are negligible and the first order phase transition easily takes place, sensitively coupled to the surrounding superflow with inertia.

To investigate its falling motion, we measured the position of the crystal at time \(t\). The position of the lowest point of the crystal \(y\) was plotted as a function of \(t^2\) in figure 3. It would be better to follow the position of the center of the mass but it was difficult to estimate it by the observation from one side. Only the position of the lowest point of the crystal was plotted because the shape change in the lower part was not as significant as the upper part. One can find a linear dependence in the early stage of roughly \(t < 0.1\) s; this means that the crystal experienced a constant acceleration motion in this period. The solid line is a linear fitting curve in the early stage and the acceleration was obtained as \((0.059 \pm 0.003)g\) from the fitting, where \(g\) is the acceleration due to gravity \(g = 9.8\) m s\(^{-2}\). In the later stage, it deviated from the linear dependence and a dissipation mechanism set in. The critical time \(t_c\) at which the deviation occurred was roughly estimated as \(t_c \approx 0.1\) s and the falling velocity at \(t_c\) was about 0.06 m s\(^{-1}\). Accurate estimation of \(t_c\) was difficult because the deviation from the line was so gradual.

Let us consider a fall of an object of mass \(m\), volume \(V\) and density \(\rho = m/V\) in a viscosity free and incompressible fluid of density \(\rho_f\). The equation of motion is
where \( M \) and \( f \) are the effective mass and the effective gravity force, respectively, and \( a \) is the acceleration. The effective force is reduced from the raw gravity value \( mg \) by the buoyancy as

\[
f = mg - \rho \Delta \rho \rho g V g = \frac{\Delta \rho}{\rho} mg,
\]

where \( \Delta \rho = \rho - \rho_l \). The effective mass is enhanced to \( m + m' \) by a liquid flow surrounding the object. In the case of a spherical object, it is known that \( m' = \rho_l V / 2 \) [22], and thus

\[
M = m + m' = m \left( 1 + \frac{\rho_l}{2 \rho} \right).
\]

Therefore, the acceleration of the spherical object can be obtained as

\[
a = \frac{\Delta \rho}{\rho} \rho g = 0.069 g,
\]

using the physical parameters of solid and liquid \(^4\)He [10, 11]. This value roughly agrees with the observed acceleration of a \(^4\)He crystal in superfluid with an accuracy of about 15%. The actual crystal shape was far from a sphere and a better estimation of the effective mass would improve the agreement. If the motion of the center of the mass of the crystal could be followed, it too might improve the agreement.

This reasonable agreement indicates that the falling was not greatly influenced by the phase transition although the drastic shape change was observed during the falling. This is in contrast to a ‘falling’ of \(^4\)He crystals in superfluid attached to a vertical wall or a ‘rising’ of superfluid droplets included in a host \(^4\)He crystal at higher temperatures [23, 24]. In these cases, the actual movement of \(^4\)He atoms in the crystals never took place and the ‘falling’ or ‘rising’ was just a pretense. The motions of the crystals or the droplets are a complete consequence of the phase transition, namely, melting in the upper part, and crystallization in the lower part induced by the gravitational energy.
In the later stage of falling as in figures 1(m–o), the crystal shape became stationary and consequently the superflow around the crystal had to be tangential to its surface in the frame moving with crystal. Kelvin–Helmholtz instability was predicted to occur on rough surfaces with tangential flows above 0.04 m s\(^{-1}\) under the gravity acceleration of g [25]. The facets in the lower part, however, were clearly flat and showed no sign of the instability. Facets were found to be robust against the tangential flow, probably because their mobility was not large enough to exhibit the instability. The rough surface in the upper part was not perfectly flat but had an undulation on a scale of 1 mm; this was possibly a manifestation of the Kelvin–Helmholtz instability. Another possible origin for the undulation was a sub-boundary or a stacking fault, which create grooves in the liquid–solid interface because of the necessary local equilibrium of surface tensions [26].

We next describe a disturbance of the falling by a tungsten needle on the fall path as shown in figure 4. This was performed in a different cell. A transducer for the nucleation was installed further up in the cell and is not visible. The diameter of the needle was 100 \(\mu\)m and the tip was sharpened by chemical etching. This \(^4\)He crystal happened to fall with its c-facet nearly horizontal at 0.4 K. Its general features were common to the oblique crystal in figure 1. The

**Figure 4.** Falling of a \(^4\)He crystal disturbed by a needle at 0.4 K. Time is indicated in each frame. The crystal was slightly hooked by the needle, drastically deformed and landed on the bottom.
lower and side surfaces were perfectly faceted and the upper surface was rough. It was the combination of growth and melting shape due to the upward superflow seen in figure 4(a). It passed near the needle with its edge slightly hooked by the tip as in figures 4(b–d). The needle caused the crystal to become slightly tilted and greatly altered in shape, so that it landed on the bottom at a position further left than the original as in figures 4(e–t). After contact with the needle, rough surfaces appeared on the side. A video clip showing the entire process in figure 4 is provided as supplementary data (available from stacks.stacks.iop.org/njp/16/113022/mmedia).

Images before and at the point of contact with the needle are shown in figures 5(a) and (b), respectively, where the influence of the needle can be seen in detail. Before the contact, the crystal had a horizontal c-facet and had reached its stationary shape. Upon contact, the facet edge protruded to the needle ahead of the c-facet which had not yet reached the position of the needle. This protrusion indicates the existence of a long-range attractive interaction between the surface of the falling crystal and the needle. Surprisingly, not only the surface closest to the needle but also the other side far from the needle protruded almost simultaneously. The crystal deformation was anomalously extended and the rim around the c-facet protruded at the moment of contact. Enlarged images in the vicinity of the tip are shown in figures 5(c–f). In figure 5, the black arrow represents the needle and the blue arrow the protrusion at the far side. The vague line parallel to the c-facet was the protruding rim and is indicated by the red arrows. The rim protruded at a velocity of about 0.15 m s$^{-1}$, significantly faster than the falling velocity of 0.05 m s$^{-1}$.

Figure 5. Enlarged images of the $^4$He crystal around the contact point with the needle in figure 4. Overall shapes before and at the contact point are shown in a and b. Enlarged images around the needle are shown in c–f for every 1 ms. Before the contact, the rim of the c-facet was drawn and stretched by the needle. The deformation was anomalously large and extended to the far side of the facet edge. The black, blue and red arrows represent the needle, the protrusion at the far side and the protruding rim, respectively.
Before the contact, the $^4$He crystal was stationary and the surrounding superflow had to be tangential to the crystal surface in the frame moving with crystal. When the crystal approached the needle, it disturbed the nearby superflow profile. The superfluid had to detour around the needle because the needle surface set a strict boundary condition on the superflow. This disturbed superflow is likely to be the origin of the attractive interaction, because the flow can be a source of the long-range force owing to its non-local nature and because crystal shapes are sensitive to the surrounding superflow as demonstrated in figure 1.

Since details of the superflow profile are not known, the true mechanism of the interaction is not clear at present. Even more unanticipated is the extended deformation around the rim of the facet induced by the local disturbance of the needle. The flow field around the crystal makes the velocity dependent pressure field via the Bernoulli effect which could be the origin of the deformation. In the laboratory frame, a downward flow may exist below the crystal, and when the crystal was hooked by the needle, the downward flow could not abruptly stop due to its inertia. It could turn into the outgoing flow perpendicular to the crystal surface and possibly induce the melting in the lower part. If the melting was larger in the central part reflecting the surrounding flow field, the resulting crystal shape may appear as in figure 5(b). Or, a new type of instability may be the other possible origin of the deformation. A simulation study of the superflow profile, which takes the crystallization process into account on an equal footing, would be needed to explain the unanticipated behavior of the falling $^4$He crystal. It would help to conclude how the tangential and/or the perpendicular flows induced the drastic shape change.

Our finding will also shed light on the hydrodynamic instability of quantum fluids which have recently been actively discussed in the field of Bose-condensed systems [1–5].

4. Summary

Falling $^4$He crystals in superfluid were demonstrated to be a rare example of a non-equilibrium system in which mass flow strongly influenced the first order phase transition resulting in a drastic change in crystal shape. Due to the coupling between the surrounding superflow and crystallization and the high anisotropy in the crystallization rate, falling $^4$He crystals showed a rich variety of shapes. The lower surface was faceted and the upper surface was rough when subjected to a unidirectional upward superflow. When the falling was disturbed by a needle, the rim of the facet protruded in advance of contact with the needle. The anomalously extended deformation may have been induced by the disturbed superflow, but this has not yet been clarified.

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References

[1] Tsubota M 2013 Hydrodynamic instability and turbulence in quantum fluids J. Low Temp. Phys. 171 571
[2] Kadokura T, Aoi T, Sasaki K, Kishimoto T and Saito H 2012 Rayleigh-Taylor instability in a two-component Bose-Einstein condensate with rotational symmetry Phys. Rev. A 85 013602
[3] Takeuchi H, Suzuki N, Kasamatsu K, Saito H and Tsubota M 2010 Quantum Kelvin-Helmholtz Instability in phase-separated two-component Bose-Einstein condensates Phys. Rev. B 81 094517
[4] Abe H, Ueda T, Morikawa M, Saitoh Y, Nomura R and Okuda Y 2007 Faraday instability of superfluid surface Phys. Rev. E 76 046305
[5] Engels P, Atherton C and Hoefer M A 2007 Phys. Rev. Lett. 98 095301
[6] Teshigawara R and Onuki A 2008 Droplet evaporation in one-component fluids: dynamic van der Waals theory Europhys. Lett. 84 36003
[7] Onuki A 2007 Dynamic van der Waals theory Phys. Rev. E 75 036304
[8] Oono Y and Paniconi M 1998 Steady State Thermodynamics Prog. Theor. Phys. Suppl. 130 29
[9] Sasa S and Tasaki H 2007 Steady State Thermodynamics J. Phys. Soc. Jpn. 77 111009
[10] Babkin A V, Kopeliovich D B and Parshin A Y 1985 An experimental investigation of faceting phase transitions in 4He crystals Sov. Phys. JETP 62 1322
[11] Elbaum M and Wettlaufer J S 1993 Relation of growth and equilibrium crystal shapes Phys. Rev. E 48 3180
[12] Maruyama M, Kuribayashi N, Kawabata K and Wettlaufer J S 2000 Shocks and curvature dynamics: a test of global kinetic faceting in crystals Phys. Rev. Lett. 85 2545
[13] Keshishev K O, Parshin A Y and Babkin A V 1981 Crystallization waves in 4He Sov. Phys. JETP 53 362
[14] Rolley E, Chevalier E, Guthmann C and Balibar S 1994 The stepped surfaces of helium-4 crystals Phys. Rev. Lett. 72 872
[15] Landau L D and Lifshitz E M Fluid Mechanics (Oxford: Pergamon)
[16] Takahashi T, Nomura R and Okuda Y 2012 Creation and annihilation of 4He negative crystals J. Phys.: Conf. Ser. 400 012070
[17] Yoneyama K, Nomura R and Okuda Y 2004 Dynamics and morphology of superfluid bubbles in 4He quantum crystals Phys. Rev. E 70 021606
Nomura R, Yoneyama K and Okuda Y 2006 Phys. Rev. E 73 049901 (erratum)
[18] Uwaha M and Nozières P 1986 Flow-induced instabilities at the superfluid-solid interface of 4He J. Phys. France 47 263
[19] Sasaki S, Caupin F and Balibar S 2008 Optical observation of disorder in solid helium 4 J. Low Temp. Phys. 153 43