Asymmetry in melting and growth relaxation of $^4$He crystals in superfluid after manipulation by acoustic radiation pressure

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Abstract

The relaxation dynamics of the crystal–superfluid interface of $^4$He after deformation induced by acoustic radiation pressure was investigated for various crystal orientations. The melting relaxation after growth was approximately 10 times slower than the growth relaxation after melting for vicinal surfaces and facets, while both relaxation times were consistent with each other for rough surfaces. The asymmetry in the time constant between the melting and growth of vicinal surfaces and facets can be qualitatively explained as the effect of superflow induced by local rapid interface motion, such as a quick rounding of facet edges of the $^4$He crystal. Rough surfaces move more isotropically and no significant local rapid interface motion is induced; therefore, their relaxation is likely to be symmetric with a minimal effect of superflow. While the growth relaxation was simply back to the initial shape in a single stage, the melting relaxation was much more complex with multiple stages and the exhibition of various anomalous shapes depending on temperature. Anomalous shapes such as needle-like shapes during melting have a larger curvature and higher energy and thus should have disappeared more quickly than the growth shape with a smaller curvature, but they were considerably stable and disappeared slowly. This counter-intuitive asymmetry suggests the significant role of superflow in the relaxation process.

1. Introduction

How the growth and melting of crystals differ from one another is a fundamental question. Although it seems reasonable to expect that both processes are not very different and proceed at a similar rate, it has been reported that melting is faster than growth [1–3], when an intrinsic mobility of the interface controls the rate of the phase transitions. However, this is not a comparison of the melting and growth velocity of the same interface; the melting process is dominated by a fast moving interface, the rough surfaces, and the growth process is dominated by a slow interface, the facets [4, 5]. The melting shape of crystals is generally determined and formed by the rough surfaces, whereas the growth shape is generally determined by the facets. Thus, it is not easy to compare the melting and growth velocity of a particular interface in crystals where the interface mobility is inherently anisotropic, because no manipulation method by which the melting and growth of the interface can be selectively driven in a desired orientation has been known.

$^4$He quantum crystals in superfluid is the exceptional material to be manipulated by small driving forces such as heat, gravity, and flow [6–13], because they grow extremely fast at low temperatures and play a very important role in understanding the fundamental aspects of crystal growth physics [14–17]. In previous reports, we have demonstrated that acoustic radiation pressure, which is the small second-order acoustic force usually not relevant to the crystallization process [18, 19], induces both growth and melting of $^4$He crystals and is a very useful tool to manipulate the interface in a desired orientation, even to a large extent [17, 20–22].
In this work, growth or melting were driven by acoustic radiation pressure, and the relaxation process after the acoustic manipulation pulse was investigated for various crystal orientations. Melting relaxation after growth was determined to be much slower than growth relaxation after melting for vicinal surfaces and $c$-facets and thus the interface motion was asymmetric in the moving directions. The melting relaxation exhibited various shapes, such as needle-like or irregular shapes, depending on the temperature. However, this asymmetry did not show up for rough surfaces. The asymmetry and the anomalous shapes can be a reflection of the superflow profile surrounding the crystal that is induced by the anisotropic interface motion. This observation of the peculiar interfacial dynamics will facilitate understanding of not only the fundamental question of crystal growth physics but also of the hydrodynamics of Bose-condensed systems in which various instabilities have been actively discussed recently [23–29].

2. Experimental procedure

Experiments were performed in a high pressure cell that was cooled with a dilution refrigerator [20–22]. The high-pressure cell had two optical windows through which the inside could be observed. Two ultrasound transducers were installed in the upper and lower part of the cell facing each other with a separation of 10 mm. The effective diameter of each transducer was around 5 mm and the resonance frequency was 9.9 MHz. Acoustic waves were directed in the vertical direction. We used a $^4$He sample with nominal $^3$He concentration of a few hundred ppb in this report, although using ultrapure $^4$He would also be an intriguing trial as a future research.

In the metastable superfluid at a few millibar above the melting pressure, an acoustic wave pulse was applied by the upper transducer to initiate nucleation of a hexagonal-closed-packed (hcp) $^4$He seed crystal on the transducer surface [13, 21, 30]. The crystal eventually fell in the superfluid and landed on the bottom. The orientation of the crystals changed during falling, so crystals with a wide range of the orientation could be obtained from rough to vicinal surfaces after several trials of this crystal preparation procedure.

Once a crystal with the desired orientation was achieved, it was grown midway between the transducers. The crystal filled the lower space of the cell because it has a higher density than the liquid. The interface became flat in a horizontal plane due to the gravity, so the acoustic wave pulses could be applied perpendicular to the interface.

Acoustic radiation pressure induces crystallization or melting, depending on the directions of wave propagation [17, 20–22]. At sufficiently low temperatures, melting of the crystal or lowering down of the interface was induced when the acoustic wave was applied downwards to the crystal–superfluid interface from the upper transducer. When the acoustic wave was applied upwards from the lower transducer, crystallization or lifting up of the interface was induced. Therefore, acoustic waves can be used to manipulate the crystal–superfluid interface of the specified orientation, which provides a way to study the relaxation process after manipulation. The volume of the crystal was kept constant during deformation because the interface mobility is so high.

However, the direction of the acoustic radiation pressure was inverted above the inversion temperature $T_i$ around 0.6 K, in which case the acoustic wave was applied from the crystal side. Only melting was induced above $T_i$ by acoustic waves in both directions [17, 20, 21].

Below $T_n$ relaxations for both growth and melting could be examined using the two acoustic wave directions. Using the upper transducer, melting could be induced by the acoustic wave pulse. After the pulse was turned off, the crystal relaxed to the initial horizontal surface with crystal growth by gravity and/or surface energy. This relaxation after melting is referred to as the growth relaxation with a relaxation time $\tau_g$. Using the lower transducer, crystallization could be induced by the acoustic wave pulse. After the pulse was turned off, the crystal relaxed to the horizontal surface with melting. This relaxation after growth is referred to as the melting relaxation with a relaxation time $\tau_m$. Both the growth and melting relaxations were investigated acoustically and visually. To observe the relaxation of the interface, the acoustic method was used for small deformations after a short acoustic wave pulse, while the visual method was used for large deformations after a long pulse. Above $T_n$, only melting was induced by acoustic waves in both directions, and thus only the growth relaxation by the acoustic waves could be examined; therefore, $\tau_g$ and $\tau_m$ could not be determined separately, but only $\tau_g$.

For a small deformation by a short acoustic pulse, the displacement of the interface induced by the acoustic wave pulse was 200 µm or less and the interface relaxed to the equilibrium position within a few milliseconds after the pulse in the fastest case. To monitor such a small displacement and fast relaxation process, the interface was manipulated and monitored acoustically. A crystal was grown or melted by acoustic radiation pressure with a manipulation pulse and it then relaxed to the equilibrium position by gravity. The position of the interface was determined by a series of monitoring pulses before and after the manipulation pulse. This is exactly the same method adopted in [21]. Time sequences of the acoustic pulses are schematically shown in figure 1(d). Although the displacement of the interface is not uniform, the average displacement of the interface over the active area of the transducer $h$, is related to the phase difference $\Delta \phi$, of the transmitted monitoring pulse at time $t$:
where $\omega$ is the angular frequency of the acoustic wave, and $c_c$ and $c_l$ are sound velocities in crystal and liquid. First, the initial position of the horizontal interface was determined prior to manipulation. Thereafter, a manipulation pulse with larger power (typically 150 W m$^{-2}$) and longer duration ($1 \text{ ms}$) than the monitoring pulse was sent at $t = 0$ and the interface was deformed. After manipulation, a series of monitoring pulses were sent to track the displacement of the interface. The monitoring pulses had less power (reduced by $-6 \text{ dB}$ from the manipulation pulse) and shorter duration ($5 \mu\text{s}$) than the manipulation pulse. The repetition rate of the monitoring pulses was changed depending on the time constant of the relaxation. The effect of the monitoring pulse on the interface motion was neglected for this power, pulse duration and repetition rate in the experimental temperature ranges employed.

To determine the crystal orientation, each crystal was grown up to the upper transducer and filled the space between the two transducers to determine $c_c$. The surface orientation $\theta$, measured from the $c$-axis was mostly determined from $c_c$ following the procedure described in [21] with the elastic constant for the hcp $^4\text{He}$ crystal [31]. For crystals with $\theta < 10^\circ$, it was also possible to determine $\theta$ visually during the sample growth process because clear $c$-facets appeared.

3. Results

3.1. Small deformations

Figure 1 (a) shows a typical time evolution of the average displacement $h(t)$ determined by phase evolution of the transmitted acoustic pulses for a vicinal surface after a manipulation pulse of 1 ms. The temperature was $T = 0.5 \text{ K}$ and the orientation $\theta$ was $2 \pm 1^\circ$. The interface was lifted up via crystallization by the manipulation pulse from the lower transducer and the displacement just after the manipulation pulse was approximately

$$\Delta \phi(t) = \omega \left( \frac{1}{c_c} - \frac{1}{c_l} \right) h(t),$$  \hspace{1cm} (1)
$h(0) = 40 \mu m$. After the manipulation pulse, $h(t)$ relaxed to $h = 0$ with melting. The interface was lowered via melting by the manipulation pulse from the upper transducer, and its displacement was approximately $h(0) = -100 \mu m$. After the manipulation pulse, $h(t)$ relaxed to $h = 0$ with growth. The melting relaxation after growth was approximately one order of magnitude slower than the growth relaxation after melting: $\tau_m = 0.77 s$ whereas $\tau_g = 0.089 s$. These relaxation times were obtained by fitting $h(t)$ to the exponential function $\exp(-t/\tau_{m,g})$. The two processes were quite asymmetric for the vicinal surface: $\tau_m \gg \tau_g$.

Figure 1(b) shows a typical time evolution of $h(t)$ determined in the same way as for figure 1(a) but for a rough surface. The temperature was $T = 0.725 K$ and the orientation $0 = 36 \pm 1^\circ$. The melting and growth relaxations were symmetric for the rough surface: $\tau_m \approx \tau_g \approx 0.05 s$.

Figure 1(c) is the semi-log plot of $|h(t)|$ in figures 1(a) and (b). The solid lines are the exponential fit of the relaxations for the vicinal surface. Small deviations from the simple exponential decay can be seen in an initial few points, which is a reflection of nonlinearity caused by local curvature and superflow as will be discussed later. Relaxation time of the average height was obtained experimentally for the small deformation but this does not mean that the surface really relaxed uniformly. The local relaxation dynamics can be different and dependent on the positions, although we cannot resolve it acoustically. We use $\tau_m$ and $\tau_g$ obtained from the fitting to represent the average behavior of the temperature and orientation dependences of the relaxations.

Figure 2 shows the relaxation time as a function of temperature for the vicinal surface and the rough surface. The inversion temperature for the small deformations of $4^\text{He}$ crystals in superfluid $^4\text{He}$ was approximately $T_i = 0.57 K$, as indicated by the arrow. The circles and squares below $T_i$ represent $\tau_m$ and $\tau_g$, respectively; $\tau_m \gg \tau_g$ and the asymmetry is very clear. Above $T_i$, only $\tau_g$ was measurable; the triangles and squares represent $\tau_g$ obtained by the lower and upper transducers, respectively. These results were consistent with each other, as shown in figure 2, although the directions of the pulses were opposite. The crosses represent $\tau_m$ for the rough surface and are much shorter than those for the vicinal surface, which reflects the high mobility of the rough surface.

Figure 3 shows the orientation dependence of the relaxation times at $0.5 K$. The circles and squares represent $\tau_m$ and $\tau_g$, respectively. For rough surfaces at $\theta > 10^\circ$, $\tau_m \approx \tau_g$, and thus the melting and growth relaxations were symmetric. As the surface orientation approached closer to the c-facet ($\theta \rightarrow 0^\circ$), the asymmetry became apparent; $\tau_m > \tau_g$ at $\theta < 10^\circ$, which was most apparent at $\theta = 2^\circ$. Therefore, the slow relaxation in melting than in growth is the specific feature of the vicinal surfaces.

The relaxation time $\tau$, is related to the growth (or melting) rate, $K$:

$$\tau = \frac{\rho_c \Delta \mu}{\Delta gK}$$

when the surface curvature can be neglected as a driving force. Here, $\rho_c$ and $\rho_l$ are the density of the crystal and liquid, respectively, $\Delta \rho = \rho_c - \rho_l$ and $g$ is gravity acceleration. $K$ is defined as:

$$\nu = K \Delta \mu,$$

where $\nu$ is the crystal–superfluid interface velocity and $\Delta \mu$ is the driving force of crystallization, i.e., the difference in chemical potential per unit mass between the crystal and superfluid. For the small deformations of the rough surface the surface curvature contribution was smaller than the gravity contribution as will be described in the discussion section. The measured temperature dependence of $\tau$ for rough surfaces (figure 2) agrees with $\tau$ calculated from $K(T)$ which Amrit et al measured by the sound transmission coefficients [32].
3.2. Large deformations

To observe the relaxations of a crystal after larger deformation, a longer acoustic wave pulse was applied to a vicinal surface and the crystal shape evolution was observed with a high speed video camera. For application of an acoustic wave for 50 ms from the crystal side at \( T = 0.5 \) K, a clear \( c \)-facet appeared on the top of the crystal, as shown in figures 4(a)–(i). The appearance of the facet is a general feature of the growth shape; the slow facet is caught up with the fast growth rough or vicinal surfaces, which results in enlargement of the facet on the growing crystal surface [2, 4, 5]. Therefore, although the initial horizontal surface was the vicinal surface, it quickly disappeared and the facet of the \( \theta = 0^\circ \) plane appeared during growth induced by acoustic radiation pressure. Here, we focus on the melting relaxation of the crystal shape after the pulse.

In figures 4(i)–(F), a typical time evolution of the largely deformed crystal is evident after the acoustic wave pulse was turned off. The facet edge melted very quickly by approximately 0.5 mm after just 1 ms and the crystal became bell shaped within a few milliseconds (figures 4(j) and (k)); the velocity of the surface \( v \), reached as high as \( v = 50 \) cm s\(^{-1}\). This interface motion induces a local superflow of the velocity of \( v_l = \Delta p v / \rho_l \approx 5 \) cm s\(^{-1}\) due to mass conservation. While the shoulder of the bell-shaped crystal continued to melt (figures 4(l)–(q)) and approached the original horizontal level at around 10 ms (figure 4(q)), a needle-like shape appeared in the center at 3 ms (figure 4(l)). The needle shape was very prominent at 8 ms (figure 4(p)) and became shorter at around 20 ms (figure 4(v)). The remaining pointed shape continued to exist for a longer period and disappeared at 300 ms (figure 4(F)). The melting relaxation consisted of two stages. In the first stage, the facet quickly disappeared and the bell shape was formed (figures 4(i)–(k)). In the second stage, the needle shape appeared in the center, and became gradually shorter while relaxing to the horizontal shape (figures 4(l)–(F)). The time constant of the second stage is of the same order of \( \tau_m \) for the small deformation case with a vicinal surface. A video clip of the entire process shown in figure 4 is provided as supplementary data.

By applying the acoustic wave pulse from the superfluid side by the upper transducer, the growth relaxation after the large melting deformation was observed. Figures 5(a)–(k) show the crystal was melted by the acoustic wave pulse for 30 ms. A \( c \)-facet was observed during the manipulation pulse, even for this melting shape, but it was not as stable as the growth shape in figure 4 and it fluctuated during the pulse. After the pulse was turned off, the crystal relaxed to the horizontal surface within 10 ms via crystal growth (figures 5(k)–(p)). The growth relaxation was smooth, occurring in a single stage, and was much faster than the melting relaxation shown in figure 4. No anomalous shapes, such as the needle-like shape, were observed during the growth relaxation and their curvature was much smaller than the needle shape. A video clip of the entire process shown in figure 5 is provided as supplementary data.

Although the growth relaxation was rather simple with only a single stage, even at lower temperatures, the melting relaxation was much slower and more complex with an intriguing temperature dependence, and the first and second stages. Figure 6 shows melting relaxations for another crystal every 1 ms following the acoustic wave pulse at 0.5, 0.45, 0.4, 0.3 and 0.2 K. At 0.5 K, the relaxation was similar to that in figure 4; the facets quickly relaxed to the bell shape in the first stage and the needle shape formed in the center at the second stage. At lower temperatures, the crystal shape became more irregular during the relaxation. At 0.45 K, the bell shape appeared at around 3 ms, but it was not as symmetric as at 0.5 K. The final needle-like shape was thicker at 6 ms. At 0.4 K, a small bump appeared on the side of the bell shape at 2 ms, and the crystal shape was no more needle-like but was much thicker at 6 ms. At 0.3 and 0.2 K, the bell shape did not appear at 2 ms, but instead the shape was more finger-like and irregular. These irregular patterns appeared in the first stage of the relaxation process.

\textbf{Figure 3.} Crystal orientation dependence of the relaxation time after small deformations at \( T = 0.5 \) K. Circles and squares represent the melting and growth relaxation times, (\( \tau_m \) and \( \tau_g \)), respectively. The horizontal bars represent the errors in orientation. Dashed lines show the general trends.
within approximately 5 ms, and thereafter relaxed to the horizontal place within a longer relaxation time of several hundreds of milliseconds in the second stage. The maximum interface velocity was observed at 0.4 and 0.3 K between 2 and 3 ms and it reached $v_{200} \approx 200 \text{ cm s}^{-1}$. The induced superflow velocity was $v_{20} \approx 20 \text{ cm s}^{-1}$.

4. Discussion

The driving of growth and melting for a particular surface of $^4$He crystals in superfluid was successfully demonstrated. It was determined that melting was much slower than growth for vicinal surfaces and facets of $^4$He crystals at low temperatures. This is contrary to the conventional knowledge detailed in the introduction. Furthermore, the melting relaxation after large deformation progressed in two stages with very complicated behavior, whereas the growth relaxation progressed in a single stage with simple behavior. However, asymmetry in the two processes for rough surfaces was not observed at all, which is consistent with the expectation that the interface mobility is symmetric in a linear response regime.

The extraordinary asymmetric relaxations are very apparent in the large deformation case. The high curvature state of the needle shape appeared in the melting relaxation in figure 4. In the growth relaxation, such

![Figure 4. High-speed camera images of crystallization by acoustic wave pulse from the crystal side (a)–(i) and melting relaxation after the pulse (j)–(F) for a vicinal surface at $T = 0.5$ K. The width of each frame is 9.5 mm.](image-url)
anomalous shapes did not appear and the curvature was much lower in figure 5. Because the displacements in both relaxations were comparable and the gravitational driving forces were on the same order, the high curvature state was a higher energy state and thus should be unstable and should relax more quickly than the lower curvature surface. However, what we visually demonstrated was that the high curvature state in figure 4 relaxed much slower than the low curvature state in figure 5. This is opposite to the above mentioned expectation.

Figure 5. High-speed camera images of melting by acoustic wave pulse from the superfluid side (a)–(k) and growth relaxation after the pulse (l)–(p) for a vicinal surface at $T = 0.5$ K. The width of each frame is 10.2 mm.

Figure 6. High-speed camera images of melting relaxation after crystallization by acoustic wave pulse from the crystal side for a vicinal surface at $T = 0.5, 0.45, 0.4, 0.3$ and $0.1$ K. The width of each frame is 10 mm.
from the energetics of the curvature. To explain this unexpected asymmetry there should be a mechanism that stabilizes the crystal phase and slow down only the melting relaxation.

The flow profiles are very different in the melting and growth relaxations; therefore, this asymmetric behavior can be accounted for by the induced superflow during the relaxation. For example, the interface recedes, leaving superfluid flow behind it in the case of melting relaxation, while the interface advances towards flowing superfluid ahead of it in the case of growth relaxation. The liquid phase with flow has higher energy than the liquid at rest, so that the solid phase is more stable when it faces the liquid with flow. Therefore, when facing the flowing fluid, melting relaxation will be slower than growth relaxation.

Let us examine the chemical potential difference $\Delta \mu$, induced by gravity, surface energy associated with curvature and by the superflow [33, 34]. When the interface is lifted up with a displacement $h$, the chemical potential difference due to the gravitational energy is given as:

$$\Delta \mu_g = -\frac{\Delta \rho g h}{\eta}. \quad (4)$$

When the interface is locally curved with a curvature $C$, the chemical potential difference due to the interfacial energy is given as:

$$\Delta \mu_C = -\frac{\gamma C}{\eta}. \quad (5)$$

Here, $\gamma$ is the surface stiffness of 4He crystal. When a supercurrent is flowing in the liquid parallel to the interface at a velocity of $v_l$, the chemical potential difference is:

$$\Delta \mu_f = \frac{1}{2} v_l^2, \quad (6)$$

which is always positive and makes the crystal phase energetically favorable under superflow conditions.

Crystallization (melting) is induced when the total driving force is positive (negative), i.e.,

$$\Delta \mu = \Delta \mu_g + \Delta \mu_C + \Delta \mu_f > 0 \quad (<=) 0. \quad (7)$$

For growth relaxation, $h < 0$ and $C < 0$, and thus $\Delta \mu_g > 0$, $\Delta \mu_C > 0$ and $\Delta \mu_f > 0$. Therefore, superflow in the liquid does not help slow down the growth relaxation, but accelerates it. For melting relaxation, $h > 0$ and $C > 0$, and thus $\Delta \mu_g < 0$ and $\Delta \mu_C < 0$, but $\Delta \mu_f > 0$. Therefore, the magnitude of the total driving force $|\Delta \mu|$, is smaller in the presence of the superflow, which can cause the melting relaxation to be slower. Consequently, the presence of the superflow in the liquid phase qualitatively explains the observed asymmetry in the relaxation processes when the superflow velocity is sufficiently high.

We first discuss the large deformation case, where the effect of superflow is most evident. A typical displacement and curvature in the second stage in figure 4(a) was approximately $h \approx 0.1$ cm and $C \approx 10$ cm$^{-1}$, which corresponds to $|\Delta \mu_g| \approx |\Delta \mu_C| \approx 10$ cm$^2$ s$^{-2}$. To compensate for these with a superflow, $v_l \approx 6$ cm s$^{-1}$ is required to make $|\Delta \mu| = 0$ in equation (7). A maximum interface velocity of $v = 50$ cm s$^{-1}$ was observed in the first stage just a few milliseconds after the acoustic pulse and a superfluid velocity of approximately $v_l \approx 5$ cm s$^{-1}$ was induced, which is comparable to $6$ cm s$^{-1}$.

A schematic illustration of the crystal profile during melting relaxation is shown in figure 7. The initial superflow fields must be normal to the moving interfaces and thus radially outgoing from the facet edge when the edge was quickly rounded, as indicated by the arrows in figure 7(b). However, the overall superflow profile and its time evolution are unknown. Provided that the initial high-velocity superflow developed into an almost stationary flow with a parallel velocity to the interface in the order of $6$ cm s$^{-1}$, as shown in figure 7(c), it would cause the slow relaxation in the second stage of melting due to the smaller $|\Delta \mu|$. The quick disappearance of the facet edge (circles in figure 7(a)) in the first stage and the formation of the bell shape seem to be reasonable because the facet edge was sharp and its curvature was large. Such a large curvature shape has a high surface energy and should be unstable without the acoustic radiation pressure field: in figure 4(h), $C \approx 40$ cm$^{-1}$ and $|\Delta \mu_C| \approx 40$ cm$^2$ s$^{-2}$. Rounding of the facet edge occurred quickly after the acoustic pulse to release the surface energy, and thus the bell shape was formed with the radial high-velocity superflow normal to the shoulder of the bell shape (figure 7(b)). This initial high-velocity superflow possibly induces the subsequent anomalous shapes and the longer relaxation time in the second stage. It is noteworthy that the sharp edge or the clear facet created by the acoustic wave is a direct consequence of the high anisotropy of the interface mobility and that the anisotropy is prominent only on the crystalline orientation around the facet but not so on rough surfaces.

After the bell shape, the needle shape appeared in the center of the bell shape at $0.5$ K during the second stage of melting relaxation. The needle shape has a large curvature at the tip; therefore, it should also be in a high-energy state and $C \approx 80$ cm$^{-1}$ and $|\Delta \mu_C| \approx 80$ cm$^2$ s$^{-2}$ in figure 4. However, it is unusual that such a high-energy and unstable state appeared and was sustained for a considerably long time, on the order of $10$ ms. The origin of the needle shape is not clear, but the initial high-velocity superflow induced by rapid motion of the
interface could play a role in the formation of the needle shape if the flow developed into the profile shown in figure 7(c).

It is true that the flow needs sources and drains. The source is the facet edge which melted very quickly just after the pulse. If the emitted superflow had been directly absorbed via crystallization on the surface out of the active area of the acoustic fields, no superflow would have survived and no anomalous relaxation would have occurred. We speculate that in the transient process the flow field like figure 7(c) is realized to cause the anomalous shapes. Eventually the flow should disappear via crystallization on the non-deformed surface. At the moment it is not obvious that this hypothetical flow field was really realized.

At lower temperatures below 0.4 K, the surface became more irregular, and a finger-like shape appeared within a few milliseconds. The appearance of the irregular shape implies that some instability mechanisms set in. The critical velocity of Kelvin–Helmholtz instability was predicted to be \( v_c = 4 \text{ cm s}^{-1} \) when a superfluid flows parallel to a horizontal crystal surface \([33]\). The initial superflow had a velocity of \( v_l = 5 \text{ cm s}^{-1} \) and the maximum velocity afterward was even higher at \( v_f = 20 \text{ cm s}^{-1} \). These are comparable to or higher than \( v_c \).

Therefore, it is possible that Kelvin–Helmholtz instability occurred during the relaxation and sustained the finger-like shape. It is reasonable that the instability was more distinct at lower temperatures, where the interface mobility is higher and the superflow is induced more intensively. Conversely, the irregular shape indicates the existence of high-velocity superflow along the crystal surface in the first stage and supports the idea of slow relaxation during the second stage in the presence of superflow.

Next, we address the small deformations of the rough surfaces, where effect of the superflow is negligible. The surface stiffness and the interface mobility of the rough surface are almost isotropic. Therefore, the displacement induced by the acoustic pulse should be smooth, which reflects the spatial distribution of the manipulation pulse intensity. In contrast to the clear edge of the facet in the case of large deformation, no sharp structure should form on the surface, as illustrated in figure 8(a). After the pulse, the interface relaxes uniformly to the horizontal surface as a whole (figure 8(b)) and thus no local rapid interface motion occurs during the melting relaxation. The induced superflow velocity is estimated to be low, \( v_l \approx 0.01 \text{ cm s}^{-1} \) in figure 1(b), and \( \Delta \mu_l \approx 5 \times 10^{-5} \text{ cm}^2 \text{s}^{-2} \). Using typical values of \( \bar{h} \approx 5 \times 10^{-3} \text{ cm} \) and \( C \approx 0.2 \text{ cm}^{-1} \), \( \Delta \mu_g \approx 0.5 \text{ cm}^2 \text{s}^{-2} \) and \( \Delta \mu_C \approx -0.2 \text{ cm}^2 \text{s}^{-2} \). Therefore, \( |\Delta \mu_g| > |\Delta \mu_l| > |\Delta \mu_C| \) and the superflow has a negligible effect on the relaxation and \( \tau_m \approx \tau_g \).

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**Figure 7.** Schematic illustration of the crystal–superfluid interface during melting relaxation after large deformation. (a) A clear c-facet in the acoustic wave field. The facet edge indicated by the circles is very sharp. (b) Quick disappearance of the edge and appearance of the bell shape after the acoustic wave pulse was turned off. The concomitant local superflow was injected normal to the moving interfaces, radially outgoing from the facet edge, as indicated by the arrows. (c) The needle shape and the hypothetical afterward superflow stream lines with a velocity field parallel to the interface. If the superfluid velocity is on the order of 6 cm s\(^{-1}\), then it will cause the slow relaxation in the second stage of melting due to the smaller \( |\Delta \mu| \).
Finally, we discuss the small deformations of vicinal surfaces. The melting relaxation was slow and the time constant $\tau_m$ was a few hundred milliseconds below 0.5 K, which was the same order of magnitude as the time constant for the second stage of melting relaxation for the large deformation. It is reasonable to assume that the slow relaxation mechanism is the same as that for large deformation: an initial high-velocity superfluid was induced by a rapid interface motion and the induced superfluid velocity is much smaller than that for the facet of figure 7.

Prior to deformation, the original vicinal surface was composed of steps aligned parallel, as illustrated in figure 9(a). During the deformation, the vicinal surface on the active area of the transducer was lifted by the acoustic wave with a typical scale of 50 $\mu$m in height and 5 mm in diameter, which corresponded to an average change in the surface orientation in the order of 1°. The orientation of the vicinal surfaces is determined by the density of steps; therefore, even a change of 1° is achieved by a significant modification in the step density.

![Figure 8](image1.png)

**Figure 8.** Schematic illustration of the rough surface during melting relaxation after small deformation. (a) A smooth deformation of the rough surface in the acoustic wave field. No sharp structure should form on the surface due to its isotropic interface mobility. (b) Relaxation to the horizontal surface after the acoustic wave pulse was turned off. No local rapid interface motion occurs during the melting relaxation and the induced superfluid velocity is much smaller than that for the facet of figure 7.

![Figure 9](image2.png)

**Figure 9.** Schematic illustration of the top view of a vicinal surface during a small deformation. The lines depict the steps on the c-plane. (a) The vicinal surface before the deformation, composed of parallel steps with equal distances. (b) A deformed vicinal surface in the acoustic wave field. The deformation is achieved by bending of the steps to alter the step density at each position. (c) Step configuration with a slightly larger deformation. If such a deformation is achieved by the acoustic wave, some of the steps will form step loops by reconnections and some will be severely bent, while the deformation is small on the macroscopic scale.
Although the induced deformation of the vicinal surfaces may appear small from a macroscopic point of view, it is achieved by the excessive bending of the steps, as illustrated in figure 9(b). When the deformation is slightly larger, some of the steps are reorganized to form step loops by reconstructions to achieve the deformation shown in figure 9(c). These strongly bent steps with the high curvature and their anisotropic configurations are unstable and should relax quickly as in the high curvature edge of facet in the large deformation case, thereby inducing a concomitant superflow after the pulse is turned off. This is in contrast to the rough surfaces with almost isotropic surface stiffness and mobility, where small deformation can be achieved smoothly by the addition of atoms at each crystalline site. While relaxation for rough surfaces was within a linear regime, relaxation for vicinal surfaces can be a nonlinear regime, even for small deformations, which results in slow melting relaxation as was the case for large deformation. The static and dynamical properties of the vicinal surfaces are known to be very anisotropic [35]; therefore, such nonlinearity can possibly emerge, even with small deformations.

It is true that the acoustic method only detects the average displacement of the interface but cannot resolve such quick and local step motions. However, in figure 1(c) a few initial points of the melting relaxation for the vicinal surface deviated from the exponential decay and showed a rapid decrease in the average displacement. This deviation possibly reflects the local relaxation process and the nonlinear motion of the highly bent steps on the vicinal surface.

Let us make a comment on screw dislocations emerging on the surface. Dislocations are the source of steps on facets and sometimes cause the nonlinear crystallization due to pinning. But on the vicinal surface the steps naturally exist because the vicinal surface is composed of the aligned steps by definition. It has been known that such vicinal surfaces can be created by making a large enough $^4$He crystal in gravity whose the facet direction is slightly tilted from the horizontal plane as demonstrated in [35]. Gravity makes the surface horizontally flat and vicinal surface is exposed naturally. This is really the case for the vicinal surfaces of figures 1–3. The step configuration would be disturbed around the emerging dislocation but the average density of steps are determined by the orientation of the vicinal surface. As demonstrated in the experiment of the crystallization wave propagation [35], steps are not pinned on the vicinal surface; the crystallization and melting occurred linearly realized by the symmetric oscillation of the steps around the equilibrium position and the crystallization waves propagated without any threshold on vicinal surfaces. Steps around the emerging dislocation were not drawn in figure 9 because they do not play any important role on the dynamics of the vicinal surface. While in case of the crystallization from vapor phase, attachment of atoms on the step may not be symmetric, step motion of $^4$He crystals is symmetric and does not account for the observed asymmetry in melting and growth.

One might think that metastable facets created by screw dislocations were the origin of the anomalous shapes and the slow melting relaxations if they existed on the top of the needle or irregular shapes and slowed down the melting relaxations. However, metastable facets usually appear during growth relaxation with $\Delta \mu > 0$ or with positive overpressure [35, 36]. For melting with $\Delta \mu < 0$, facets can shrink from the edge without any threshold because the facet edge is adjacent to the vicinal or rough surfaces which have larger mobility [2, 37, 38]. In fact, the facet edge quickly shrank and the bell shape formed in figure 4(k), which can be regarded as a rough surface during the melting with $\Delta \mu < 0$ and should not be the metastable facet.

It is known that Grinfeld instability occurs in $^4$He crystal when it is uniaxially stressed [39]. Acoustic radiation pressure is not an isotropic hydrostatic pressure but is more precisely a uniaxial stress [18, 19], and thus one might think that the irregular shape was created by Grinfeld instability rather than by Kelvin–Helmholtz instability. Grinfeld instability, however, is induced when the stress is applied parallel to the crystal surface. In the present experiment, acoustic wave was applied perpendicular to the crystal surface and the applied stress at the surface mostly functioned to drive the crystallization or melting due to the high mobility of the surface. Therefore, direction of the stress was perpendicular to the ordinary direction for the Grinfeld instability and the stress accumulated in the crystal is supposed to be too small to cause the instability. It should also be noted that the irregular shape appeared only after the acoustic pulse was turned off and the stress was released. We believe that Grinfeld instability is ruled out as the cause of the irregular shape.

To draw a final conclusion regarding the asymmetric relaxation and anomalous relaxation shapes, however, requires determination of the overall profile of the superfluid field induced by the initial rapid surface motion and its subsequent time evolution. To determine the proposed mechanism, it is crucial to identify whether the superflow profile with a parallel component to the crystal surface is sustained for the time scale of the second stage, even when crystal–superfluid phase conversion can occur very quickly. The superflow velocity is so high that the quantum vortices are likely to be generated densely. Although some flow imaging techniques have been developed recently [40–44], these are difficult to apply at the very low temperatures used in the present experiments. Simulation study of the superflow would be a possible and conclusive way to determine the superflow profile, but it would not be an easy task; both the superfluid hydrodynamics and the crystal–superfluid phase transition would need to be taken into account on an equal footing. However, the phase transition is highly anisotropic in both a static and dynamic sense, and the crystallization rates have a very large
temperature dependence [14–17]. The hydrodynamics must also be examined under the strong influence of the crystal shape, which set a time dependent boundary condition. Conversely, the crystal shape is also determined by the surrounding superfluid field, which induces anisotropic crystallization and instability characteristics. Emergence of the time evolution of the crystal shape should be the result of cooperative phenomena of superflow and phase transition in the highly nonlinear regime. In this sense, the time evolution of the crystal shape itself is a crucial indication of the invisible superfluid motion that places a severe constraint on simulation studies.

5. Summary

Acoustic radiation pressure pulses were used to investigate the relaxation dynamics of the $^4$He crystal–superfluid interface for small and large deformations. In the case of a small deformation by a short pulse, the time for the melting relaxation after growth by the acoustic pulse was the same as that for growth relaxation after melting for rough surfaces. However, the melting relaxation for vicinal surfaces was approximately 10 times slower than the growth relaxation. In the case of large deformation by a long pulse, vicinal surfaces turned into the $c$-facet during the manipulation pulse, due to the low mobility of the facet. For melting relaxation at a high temperature of 0.5 K, the needle shape appeared in the center of the facet after the pulse within 10 ms. The subsequent pointed shape was stable and remained as long as 300 ms. At lower temperatures, the melting relaxation became more irregular and a finger-like shape appeared after the pulse instead of the needle shape; it was also stable and remained for a few hundred milliseconds. The melting relaxation consisted of two stages to return to the initial flat shape. The growth relaxation after melting was much simpler; relaxation occurred in a single stage to the flat shape within 10 ms, which was much faster than the melting relaxation. Considering these results, the asymmetry in the growth and melting processes of $^4$He crystals was observed for the $c$-facet and its vicinal surfaces, which are anisotropic, but was not observed with the isotropic rough surfaces.

Since the higher energy shapes with higher curvature in melting relaxed more slowly than the lower energy shapes with lower curvature in growth, the observed asymmetry is counter-intuitive. The asymmetry probably reflects the superflow profile surrounding the crystal that is induced by the anisotropic interface motion. Flowing superfluids have higher energy than those at rest; therefore, the coexisting crystal should be more stabilized and the melting relaxation should be slowed down if the superflow velocity is sufficiently high. This can also induce interfacial instabilities and be a source of the irregular shapes. However, simulation studies of the flow profile are definitely required to reach a conclusion. While the superflow is invisible, the moving interface during relaxation sets a time dependent boundary condition and has a strong influence on the superflow profile; therefore, the anomalous crystal shape itself should be a crucial indication of the time evolution of the superflow and the observed asymmetry during relaxation.

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References

[1] Weeks J D and Gilmer G H 1979 Dynamics of crystal growth Adv. Chem. Phys. 40 157
[2] Nomura R, Hensley H H, Matsushita T and Mizusaki T 1994 Crystal growth of U$_3$D$_2$, solid $^3$He J. Low Temp. Phys. 94 377
[3] Amrit J and Bossy J 1993 Mobility of the $^3$He crystal–liquid interface at the minimum of the melting curve J. Low. Temp. Phys. 92 415
[4] Elbaum M and Wettlaufer J S 1993 Relation of growth and equilibrium crystal shapes Phys. Rev. E 48 3180
[5] Maruyama M, Kuriyabashi 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
[6] Wolf P E, Edwards D O and Balibar S 1983 Measurements of the Kapitza resistance and Onsager cross-coefficient for the $^4$He crystal–superfluid interface J. Low. Temp. Phys. 51 489
[7] Saitoh Y, Ueda T, Ogasawara F, Abe H, Nomura R and Okuda Y 2006 Solid–liquid interface motion of $^4$He induced by heat pulse AIP Conf. Proc. 850 345
[8] Takahashi T, Nomura R and Okuda Y 2015 Equilibrium shape of $^4$He crystal under zero gravity below 200 mK Sci. Adv. 1 e1500825
[9] Takahashi T, Ohuchi H, Nomura R and Okuda Y 2012 $^4$He crystals in superfluid under zero gravity Phys. Rev. E 85 030601(R)
[10] Takahashi T, Ohuchi H, Nomura R and Okuda Y 2012 Ripening of splashed $^4$He crystals by acoustic waves with and without gravity New J. Phys. 14 123023
[11] Yoneyama K, Nomura R and Okuda Y 2004 Dynamics and morphology of superfluid bubbles in $^4$He quantum crystals Phys. Rev. E 70 021606
[12] Maksimov L A and Tsymbalenko V L 2002 The instability of the surface of helium crystal in a superfluid flow J. Exp. Theor. Phys. 95 455
[13] Nomura R, Yoshida T, Tachiki A and Okuda Y 2014 Falling $^4$He crystals in superfluid New J. Phys. 16 113022
[14] Balibar S, Alles H and Parshin A Ya 2005 The surface of helium crystals Rev. Mod. Phys. 77 317
[15] Balibar S and Nozières P 1994 Helium as a probe in materials science Solid State Commun. 92 19
[16] Lipson S G and Polturak E 1987 The surface of helium crystals Progress in Low Temperature Physics ed D F Brewer vol 11 (Amsterdam: North-Holland) p 127
[17] Okuda Y and Nomura R 2008 New aspects of crystal growth of solid $^4$He studied by acoustic wave J. Phys. Soc. Japan 77 111009
[18] Rayleigh L 1902 On the pressure of vibrations Phil. Mag. 5 358
[19] Sato M and Fujii T 2001 Quantum mechanical representation of acoustic streaming and acoustic radiation pressure Phys. Rev. E 64 026311
[20] Nomura R, Suzuki Y, Kimura S and Okuda Y 2003 Interface motion and nucleation of solid helium–4 induced by acoustic waves Phys. Rev. Lett. 90 075301
[21] Nomura R, Kimura S, Ogasawara F, Abe H and Okuda Y 2004 Orientation dependence of interface motion in $^4$He crystals induced by acoustic waves Phys. Rev. B 70 054516
[22] Abe H, Saitoh Y, Ueda T, Ogasawara F, Nomura R, Okuda Y and Parshin A Ya 2006 Facet growth of $^4$He crystal induced by acoustic waves J. Phys. Soc. Japan 75 023601
[23] Tsubota M 2013 Hydrodynamic instability and turbulence in quantum fluids J. Low Temp. Phys. 171 571
[24] Kadokura T, Aioi 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
[25] 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
[26] Abe H, Ueda T, Morikawa M, Saitoh Y, Nomura R and Okuda Y 2007 Faraday instability of superfluid surface Phys. Rev. E 76 046305
[27] Engels P, Atherton C and Hoefer M A 2007 Observation of Faraday waves in a Bose–Einstein condensate Phys. Rev. Lett. 98 095301
[28] Abe H, Ueda T, Morikawa M, Saitoh Y, Nomura R and Okuda Y 2007 Faraday instability of crystallization waves in $^4$He J. Phys.: Conf. Ser. 92 012157
[29] Abe H, Morikawa M, Ueda T, Nomura R and Okuda Y 2008 Instability of the solid–liquid interface of $^4$He during a large transformation J. Low Temp. Phys. 150 289
[30] Abe H, Ogasawara F, Saitoh Y, Tatara T, Kimura S, Nomura R and Okuda Y 2005 Nucleation of crystals and superfluid droplets in $^4$He induced by acoustic waves Phys. Rev. B 71 214506
[31] Crepeau R H, Heybey O, Lee D M and Strauss S A 1971 Sound propagation in hcp solid helium crystals of known orientation Phys. Rev. A 3 1162
[32] Amrit J, Legros P and Poitrenaud J 1995 Rough and vicinal surfaces of helium–4 crystals, mobility measurements J. Low Temp. Phys. 100 121
[33] Uwaha M and Nozières P 1986 Flow-induced instabilities at the superfluid–solid interface of $^4$He J. Phys. France 47 263
[34] Parshin A Y 1995 Equilibrium shape of rotating helium crystals Physica B 210 383
[35] Rolley E, Guthmann C, Chevalier E and Balibar S 1995 The static and dynamic properties of vicinal surfaces on helium–4 crystals J. Low Temp. Phys. 99 851
[36] Ruutu J P, Hakonen P J, Babkin A V, Parshin A Y, Penttilä J S, Saramaki J P and Tvalashvili G K 1996 Facet growth of $^4$He crystals at mK-temperatures Phys. Rev. Lett. 76 4187
[37] Tsepelin V, Saramaki J P, Babkin A V, Hakonen P J, Hyvonen J J, Luusalo R M, Parshin A Ya and Tvalashvili G K 1999 Elementary steps on the $^4$He crystal interface probed by $^3$He atoms Phys. Rev. Lett. 83 4804
[38] Kawaguchi Y, Ueno T, Kinoshita Y, Sasaki Y and Mizusaki T 2002 Crystal growth and melt of nuclear–ordered solid $^3$He J. Low Temp. Phys. 126 27
[39] Torii R H and Balibar S 1992 Helium crystals under stress: the grinfeld instability J. Low Temp. Phys. 89 391
[40] Yarmchuk E J, Gordon M J V and Packard R E 1979 Observation of stationary vortex arrays in rotating superfluid helium Phys. Rev. Lett. 43 214
[41] Bewley G P, Lathrop D P and Sreenivasan K R 2006 Visualization of quantized vortices Nature 441 588
[42] Gordon E B, Nishida R, Nomura R and Okuda Y 2007 The filament formation by impurities embedding into superfluid helium JETP Lett. 85 581
[43] Zmeev D E et al 2013 Excimers He2 as tracers of quantum turbulence in $^4$He in the $\Gamma$ = 0 limit Phys. Rev. Lett. 110 175303
[44] Gomez J F et al 2014 Shapes and vorticities of superfluid helium nanodroplets Science 345 906