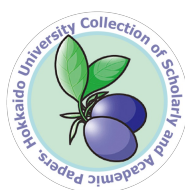




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# Anomalous Oscillation Modes of Superfluid Pendant Droplets

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Droplets should exhibit various dynamical phenomena when adhered to a surface; not all of them are realized in classical fluids. Visualization of superfluid  $^4\text{He}$  pendant droplets revealed that the droplets were horizontally translated on a flat surface, bouncing off at the corner, known as the Noether mode that reflects the translation symmetry. The droplets exhibited another mode in vertical oscillations with high amplitude that included oscillation of the droplet edge. The oscillation period remained constant even as the droplets grew, exhibiting an anomalously weak size dependence. The high mobility of the droplet edges owing to the superfluidity was a crucial factor for the appearance of these anomalous modes.

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Droplets are a common occurrence in nature and have various industrial applications [1]. They can exist freely in space, be surrounded by a medium, or be adhered to a surface. Droplets exhibit different oscillational features according to their environment and this impacts the dynamics of the system. While the oscillation of free droplets has been studied extensively, the oscillation of droplets on a substrate, whether sessile or pendant, is still under active discussion due to recent theoretical and experimental developments [2–7]. However, these discussions have mainly focused on classical fluids with viscosity, and it is unknown whether superfluid droplets on a substrate exhibit fundamentally different behavior from those of classical fluids [8–13]. Superfluid  $^4\text{He}$  has unique characteristics not present in classical fluids, such as zero viscosity, a low damping surface mode as ripplons, and complete substrate wetting [14–16]. These characteristics may alter droplet oscillation. Here we discuss the discovery of two anomalous modes of superfluid pendant droplets: the Noether mode and the anomalous (2, 0) mode, which are only realized in a superfluid.

Rayleigh calculated the oscillation frequency of a free droplet as  $\omega = 2\pi/T = \sqrt{\sigma/\rho R^3 \times l(l-1)(l+2)}$  for the modes where the distortion from the sphere is specified by the spherical harmonic function  $Y_{l,m}(\theta, \phi)$  [17,18].  $T$ ,  $\sigma$ ,  $\rho$ , and  $R$  are the oscillation period, the surface energy, the density of the liquid, and the radius of the droplet, respectively. The oscillation frequency has a degeneracy in the degree  $m$  of  $Y_{l,m}$  when the order  $l$  is the same. This equation shows that a larger droplet oscillates with a longer period. It also shows that there exists a zero frequency mode,  $\omega = 0$  for  $(l, m) = (1, 1)$ , known as the Noether mode, which is a translational mode and is trivial for free

droplets [2–4]. However, this mode is typically impossible when a droplet of a classical fluid attaches to a horizontal wall with microscopic roughness, which breaks the translational symmetry and pins the contact line [1,19]. There is a discussion about the spontaneous occurrence of the Noether mode for sessile droplets [2–4]. The question of interest is what happens in the Noether mode when a superfluid droplet adheres to a wall.

The oscillational properties of a droplet adhered to a wall have been actively discussed. Theoretical investigations have been conducted on sessile droplets to calculate the oscillation frequencies and shapes with the contact angle of the fluid to the wall  $\alpha$ , and the mobility of the contact line  $\eta$ , taken into account [2–4]. The presence of a wall lifts the degeneracy in  $m$ . It was also discovered that the modes can be identified by the same indices  $(l, m)$  as for free droplets when in the presence of a wall. This is possible because the mode frequencies of specific  $(l, m)$  change continuously as a function of  $\alpha$  and  $\eta$ , and can thus be traced. Experiments conducted on the ground and at the International Space Station confirmed this finding for sessile droplets, although the range of  $\alpha$  and  $\eta$  in the experiments was limited [5–7]. Superfluid  $^4\text{He}$  is ideal to investigate the extreme conditions of  $\alpha = 0$  and  $\eta = \infty$ , which cannot be realized in classical fluids. Because of the complete wetting of superfluid  $^4\text{He}$ ,  $\alpha = 0$ , so that the wall is always covered by a thin superfluid film. While a superfluid sessile droplet quickly spreads over the horizontal wall and thus cannot stably exist, a superfluid pendant droplet suspended from such a wall can be realized. It is important to note that the contact line cannot be uniquely defined for a pendant droplet with  $\alpha = 0$ , because the wall is coated with a preexisting thin film. Therefore, instead of the contact line, we refer to the droplet edge as that location where the thickness of the superfluid becomes lower than the resolution of the images.

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We visualized oscillating pendant droplets that were suspended from the bottom of a cup with various bottom shapes. To drive the oscillation, the spontaneous flow from a cup with an open top was utilized, which is called superfluid film flow [20–22]. As the superfluid liquid flowed into the droplet through the thin film on the cup surface, it grew on the bottom. Once it reached its critical mass, it stretched downwards, pinched off, and fell. The pendant droplet that remained on the bottom recoiled upward quickly after the pinch-off, which triggered a large amplitude oscillation. This oscillation continued until the next pinch-off and this dripping process was repeated until the cup was empty. The dripping period was mainly determined by the flow rate, which was roughly adjustable by the position of the cup from the level of the superfluid. In this Letter, we focus on the oscillation process for pendant droplets formed between two pinch-offs.

The droplets were observed in a glass Dewar flask for which the surface was silver plated to reduce thermal radiation and with a nonplated slit that allowed observation of the cup. The liquid  $^4\text{He}$  in the Dewar was cooled to the superfluid phase through evaporative cooling by pumping the vapor. The experiment was conducted at a base temperature of 1.6 K. A glass cup with a 30 mm diameter was attached to the end of a moving rod and introduced into the Dewar through a top flange by using an o-ring compression seal known as the Wilson seal. The rod was lowered to scoop the superfluid  $^4\text{He}$  from the Dewar into the cup, which was then lifted above the superfluid surface, typically several centimeters above it. This caused the superfluid in the cup to spontaneously drip from the bottom. The cup surface was coated with a commercial car

mirror-coating agent to maintain a film flow rate that was suitable for the experiment [22].

The cup was illuminated from the back of the Dewar using parallel light to visualize the droplet profile. A high-speed video camera (Photron, FASTCAM SA CGV3) was used to capture the shadowgraph image from the front. Videos were taken at a rate of 125 frames per second or higher. The image brightness was differentiated and then binarized using an appropriate threshold to detect the droplet profile. The profile resolution was a few pixels or approximately 0.1 mm.

Cups with cylindrical columns attached to their bottoms were employed to observe the oscillations of superfluid pendant droplets when suspended from walls of different shapes. The flat bottom columns used were made of brass and were 1 to 6 mm in diameter ( $d$ ) and approximately 7 mm long. The droplet oscillations on the bottom of these columns were observed in addition to those on the semi-spherical bottoms of bare glass cups without a column, as illustrated in Figs. 1(a) and 1(b).

For the droplet motion on a wide column, a peculiar translational mode was occasionally observed on a sufficiently wide flat bottom. Figure 1(c) shows that for  $d = 5$  mm, a pendant droplet traveled on the flat surface and reflected at the corner, exhibiting an oscillation in the horizontal direction. See the movie file SMmovie1 [23]. This is a (1, 1) mode or the Noether mode, which has been theoretically hypothesized but has never been observed in classical fluids. The Noether mode is the horizontal movement of a droplet at zero frequency that results from the translation symmetry of the system [2–4]. The Noether mode can only occur when the contact line or the edge of a

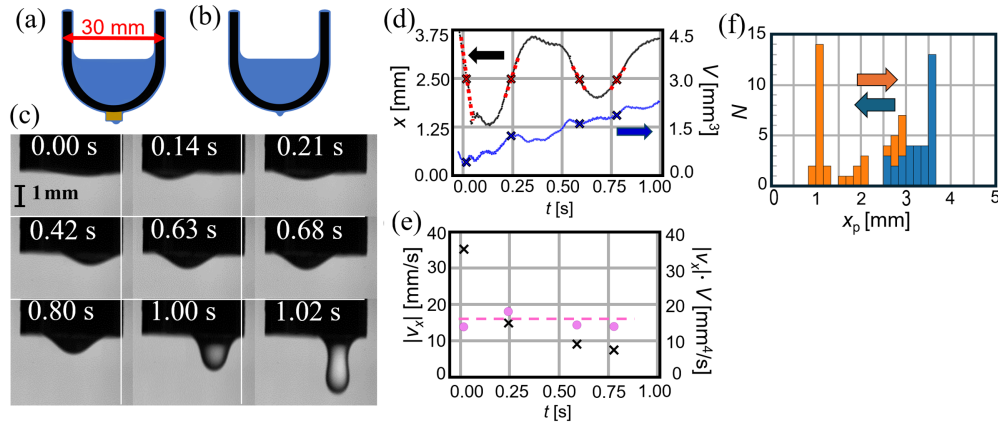


FIG. 1. (a),(b) Illustrations of the superfluid pendant droplets dripping due to the film flow from glass cups (a) with and (b) without a column on the bottom. (c) Horizontal oscillation mode or Noether mode for a superfluid  $^4\text{He}$  droplet on a wide column with  $d = 5$  mm. Time is indicated in each frame. (d) The horizontal position of the lowest point of the droplet  $x$  and its volume  $V$  in (c) as a function of time. The arrows indicate the relevant axis. The crosses show the point where the droplet passed the column center at  $x = 2.5$  mm. The horizontal velocity  $v_x$  was estimated from the time derivative around these crosses as the dotted lines. (e) Magnitude of  $v_x$  (crosses) and  $|v_x| \cdot V$  (circles) as a function of time. As  $V$  increased over time,  $|v_x|$  decreased. However,  $|v_x| \cdot V$  nearly remained constant, representing the conservation of momentum. The horizontal line is the guide for the eye. (f) Histogram showing the direction of travel based on the horizontal position of the former pinch-off  $x_p$ . Arrows indicate travel direction.

droplet can move freely. Such high mobility is not feasible in a realistic system of a classical fluid, so that this mode appears to be almost virtual. However, pendant droplets of the superfluid had sufficiently high edge mobility that the virtual Noether mode on the flat bottom of a wider column was realized. This phenomenon can only occur in superfluid  $^4\text{He}$  because the surface is covered with a thin film of the superfluid, which prevents the edge from getting pinned. In contrast, the edge of a classical fluid droplet is pinned by the microscopic roughness of the surface [1,19].

The horizontal motion of the droplet and its reflection upon meeting the corner indicate that it is the Noether mode. If the droplet moved only because the bottom surface was inclined, then it would not be classified as the Noether mode. Such motion on an inclined surface is unidirectional towards the lower end and can occur in classical fluids. However, the horizontal oscillation of the superfluid droplet indicates that it can move in any horizontal direction due to the translational symmetry of the flat bottom. When the droplet meets the corner, which breaks the translation symmetry, the droplet is reflected. This is shown in Fig. 1(d) presenting the horizontal position of the droplet's lowest point  $x(t)$  and volume  $V(t)$  from Fig. 1(c) plotted against time  $t$ .  $V$  was estimated from the droplet images assuming its cylindrical symmetry.  $V$  increased nearly proportional to  $t$  due to the film flow. In Fig. 1(e), the magnitude velocity  $v_x$  of the droplet and  $|v_x| \cdot V$ , which is proportional to its momentum, are shown at the moment indicated by the crosses in Fig. 1(d). As the droplet grew,  $|v_x|$  decreased, but the momentum was nearly conserved. This suggests that the inertia imparted from the constant flow feeding the droplet from the film is not the major source of the horizontal motion and supports the realization of the Noether mode.

How the horizontal mode is excited and how the initial direction of travel is chosen are of particular interest. Observations have shown that when a droplet begins to move to the left (right), this was due to the former pinch-off that occurred in the vicinity of the right (left) corner of the column bottom. A histogram of the initial direction as a function of the former pinch-off position on the bottom  $x_p$  is shown in Fig. 1(f);  $x_p$  affected the direction of the subsequent travel indicated by the arrows. The presence of the corner caused an asymmetric recoil after pinch-off, which drove the movement in the opposite direction. When a droplet pinched off around the middle of the bottom, the horizontal motion of the next droplet was not significant. See the movie file SMmovie2 [23]. Observation of the droplets could only be made from one side; therefore, we were unable to distinguish whether the droplets stayed around the middle position or oscillated along the line of sight. However, the existence of the Noether mode can be clearly seen. It may seem as if the droplet is circulating along the corner in Fig. 1(c), whereas the result of Fig. 1(f)

indicates a repulsive force between the droplet and the corner. Therefore, it can be concluded that the droplet is oscillating rather than circulating, even when observed from only one side. Although the spontaneous horizontal translation due to instability was mentioned in Refs. [2] and [3], our findings support that the mode needs an initial drive to be excited for superfluid pendant droplets, as recently mentioned in Ref. [4].

At a higher flow rate, an alternating horizontal motion was observed after each droplet fell. A droplet pinched off at one corner, traveled to the other direction via the Noether mode, pinched off at the other corner, and repeated this alternately. See the movie file SMmovie3 [23].

The oscillational behavior of a pendant droplet on a bare glass cup that provides a smoothly curved bottom surface is discussed next. No horizontal mode occurred, because the droplet formed at the lowest position of the surface, showing clear results not influenced by the Noether mode. When the droplet was pinched off, the droplet that remained on the bottom quickly recoiled upwards, which caused a large amplitude oscillation. The oscillation then continued with a high amplitude that was comparable to the droplet size, as shown in Fig. 2(a). The droplet size gradually increased as the superfluid continued to flow into the droplet via the film flow. The oscillation amplitude remained mostly unchanged during this growth, which indicates that the oscillation was minimally damped by the superfluidity. See the movie file SMmovie4 [23].

The oscillation of the droplet caused a large-scale deformation that occurred throughout the entire body of the droplet and not just in the lower part. As shown in Figs. 2(b) and 2(c), the edge of the droplet on the bottom surface oscillated simultaneously with a high amplitude, which indicated high edge mobility. This strongly contrasts the oscillation on narrow columns in Fig. 2(d) as explained

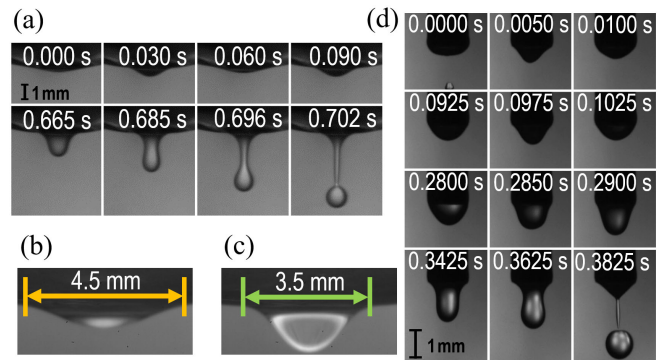


FIG. 2. (a) Oscillation and pinch-off of a superfluid  $^4\text{He}$  pendant droplet on the semispherical bottom of a bare glass cup. While suspended from the bottom, it oscillates in high amplitude. (b),(c) Expanded droplet oscillation images showing the cooperative edge motion. (d) Oscillation and pinch-off of a droplet on a narrow column with  $d = 1.5$  mm. The droplet edge was pinned at the corner of the column during the oscillation. Times are indicated in each figure.

later. The smallness of the damping is also due to the superfluidity; the edge motion is inevitably accompanied by a significant nearby flow, which should cause heavy damping of the oscillation if the fluid is viscous. However, superfluids are free from such damping; therefore, the observed large-scale and undamped oscillation is a unique behavior for superfluid pendant droplets. The image of the oscillation shape indicates that it is a quadrupole (2, 0) mode [2,3,7]. The oscillation period for the (2, 0) mode is calculated to be 0.05 s using Rayleigh's equation when the radius of the droplet is 1 mm. This value is approximately the same as the observed period of 0.08 s. The uncertain radius for the high amplitude oscillation makes it difficult to estimate an accurate value, but this rough agreement supports the mode identification. We previously reported that high-amplitude oscillations dominated the dripping process, which discretized the dripping period according to the number of oscillations [24]. This oscillation suppressed the chaotic motion that is typically observed in classical fluids [25,26].

Another point of interest for the oscillation on the bare cup is the weakly size-dependent oscillation period. Even though the droplet grows on the bottom, the oscillation period remains almost constant from just after the pinch-off to the next pinch-off, as shown in Fig. 3(a). The vertical length of the droplet  $L(t)$ , which is measured by the distance between the lowest point of the droplet and the bottom surface, is shown here as a function of  $t$ . As the superfluid flows into the droplet, the droplet size increases, and thus its overall position moves lower. The oscillation period  $T_o$  was nearly time independent for the bare glass cup. Flow rate dependence is shown for the bare glass cup in Fig. 3(b). While the flow rate increased by about 2.3 times from upper to lower plots,  $L(t)$  had the similar oscillation period indicating that  $T_o$  is independent on the flow rate, too. To see the independence clearly, the time origins were shifted to align the peak position at the time of the dashed line.  $T_o(t)$  and  $V(t)$  for the glass cup in Fig. 3(a) are plotted as the circle and crosses in Fig. 3(c). The final volume was about 2 times larger than the initial volume. According to Rayleigh's equation, the oscillation period should have increased by 1.4 times during the oscillation. The observed increase was at most 15%, showing the weak size dependence. However, a longer period is typically expected for a larger droplet. Such an increase in the oscillation period has been observed in similar droplet systems, such as water dripping from a faucet [27,28]. The weakly size-dependent period observed is thus counterintuitive and unexpected. Therefore, we refer to this oscillation mode as the anomalous (2, 0) mode.

Finally, we address the dependence of the oscillational property on  $d$ . When  $d$  is less than 2.5 mm, which is narrower than a typical droplet width observed on the bare cup, the droplet edge is pinned at the corner between the bottom and side of the column, as shown in Fig. 2(d). See

the movie file SMmovie5 [23]. The corner acts as a pinning site because the shape of the droplet must change significantly to adjust to the boundary condition if the edge goes beyond the corner. This pinning is thus geometrical.  $L(t)$ ,  $T_o(t)$ , and  $V(t)$  are shown in Figs. 3(a) and 3(c) for  $d = 1.5$  mm. While the oscillation amplitude was large and undamped, as on the bare cup, the oscillation period was size-dependent and became longer over time, as observed in water droplets from a faucet [27,28]. Therefore, the observed mode was not anomalous, and is thus referred to as the normal (2, 0) mode. The pinning suppressed the occurrence of the anomalous (2, 0) mode. The period increase was almost proportional to  $t$ , while a  $t^{1/3}$  dependence was reported for water dripping [27,28].

When  $d$  is wider than 3 mm, the bottom surface allows the edge to move freely, which results in a weakly size-dependent oscillation period similar to that for the bare cup; an anomalous (2, 0) mode is observed.  $L(t)$  is shown in Fig. 3(a) for  $d = 3$  mm. The anomalous oscillation was observed to be crucially dependent on the free motion of the droplet edge, which allowed the entire body of the droplet to oscillate at high amplitude. For the intermediate case of  $d = 2.75$  mm, the oscillation appeared irregular, as shown in Fig. 3(a). Fast Fourier transform (FFT) spectra of

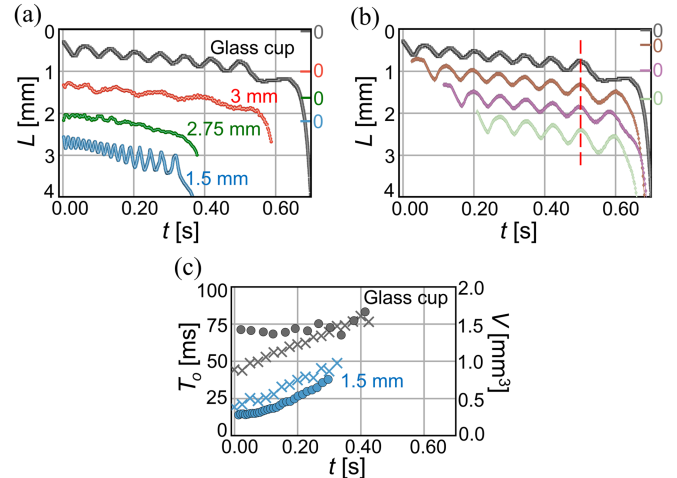


FIG. 3. (a),(b) Time evolution of the vertical length  $L$  of pendant droplets from the bottom between two pinch-offs. The origins of  $L$  are indicated on the right-hand axis. Bottom shape dependence is shown in (a). The oscillation period  $T_o$  was nearly time-independent for the bare glass cup but was dependent for the narrow columns. Flow rate dependence is shown for the bare glass cup in (b).  $T_o$  remained constant even as the rate increased by about 2.3 times from upper to lower plots. The time origins were shifted to align the peak position at the time of the dashed line. (c)  $T_o$  (circles) and the droplet volume  $V$  (crosses) as a function of time for the bare cup and the column with  $d = 1.5$  mm in (a).  $V$  increased due to the film flow.  $T_o$  was volume dependent for the column but nearly independent for the bare glass cup.

$L(t)$  for various bottom shapes is shown in the Supplemental Material [23].

In conclusion, an experiment was conducted to visualize the behavior of superfluid pendant droplets that were suspended from the bottom of a glass cup with various bottom shapes. An anomalous horizontal oscillational motion, known as the Noether mode, was observed for wide columns. This was due to the negligible pinning of the droplet edge, which resulted in the translational symmetry of the system. Another anomalous mode was also observed where the entire droplet, including its edge, oscillated at a high amplitude with little damping for the bare cup and wide columns. The oscillation period remained unexpectedly constant over time, regardless of the size of the droplet. However, for narrow columns, the droplet edge was pinned by the corner, and the oscillation period increased as the droplet size increased, which is consistent with the behavior of classical fluid droplets. It should be noted that these anomalous oscillation modes are unique to superfluid  $^4\text{He}$  because it is the only fluid that has high mobility at the droplet edge.

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