

Coalescence of Crystalline Drops

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We present the first experimental analysis of drop coalescence in a case where the dynamics is not governed by viscous dissipation in the bulk nor by the inertia of the fluid flow, only by the geometry and mobility of surfaces. We found such a situation in the physics of ^3He crystals near 0.32 K where the latent heat of crystallization vanishes. Two crystalline drops of ^3He coalesce if their crystalline orientations are identical: a neck forms after the contact at time $t = 0$, and the shape evolves towards that of one convex crystal by local growth and melting in a fraction of a second. We have found that the neck radius initially increases as $t^{1/3}$, as predicted by Maris. This behavior is also expected for superfluid drops. It is clearly distinguished from the logarithmic behavior and from the $t^{1/2}$ power law which have been predicted by Eggers *et al.* in more usual situations.

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When two liquid drops come into contact, a neck forms whose radius R_n quickly grows as a function of time t . This is the beginning of coalescence, whose study is motivated not only by important applications (printing, sintering, etc.) but also by fundamental considerations. Coalescence is an example of singularity in fluid dynamics for which at least three different scaling laws have been proposed. Hopper [1] and Eggers [2] showed that, at early times, the dynamics should be governed by viscous dissipation in the bulk fluids, so that R_n should grow as $t \ln(\gamma t / \eta R)$, where R is the common radius of the two coalescing drops before contact, γ is the surface tension, and η the viscosity of the liquid inside the drops. Some experimental evidence for this viscous regime has been found by Aarts *et al.* [3]. Eggers *et al.* also predicted a crossover to an inertial regime where $R_n \propto t^{1/2}$, when the Reynolds number is much larger than 1. The latter behavior is under active experimental study [4–7].

Maris [8] considered a different possible regime where dissipation only occurs at surfaces that move with a finite mobility. This should be the case for superfluids where the bulk viscosity is zero, and for which Maris predicted a $t^{1/3}$ behavior. He further predicted that the same $t^{1/3}$ power law should describe the coalescence of helium crystals in their surrounding liquid phase; this is because the physics is the same, dissipation occurring only at surfaces. In the latter case, Maris obtained the precise equation

$$R_n^3 = \frac{6\pi k \gamma}{\kappa \rho_C} t, \quad (1)$$

where γ is the surface tension of the crystal, ρ_C is the crystal density, and $\kappa = 2/R$ is the common initial curvature of the two crystalline drops coming into contact; the “growth coefficient” or “surface mobility” k characterizes the dissipation at the crystal surface [9–12]. It relates the growth velocity v to the difference in chemical

potential per unit mass $\Delta\mu$ between the crystal and the liquid through the relation $v = k\Delta\mu = k(\mu_L - \mu_C)$. In this particular case where the temperature is homogeneous, $\Delta\mu$ is proportional to the pressure difference across the moving surface, i.e., to the product $\gamma\kappa$. If one tried to verify this behavior with two superfluid helium drops, the coalescence would be very fast and difficult to study. Even with ^4He crystals, the growth dynamics is rather fast. Maris had anticipated that ^3He crystals could be a good system to study near $T_{\min} = 0.32$ K, the minimum in the melting curve where the latent heat vanishes. He proposed that one somehow levitates two crystalline drops and forces them to come into contact. For this to work, the orientation of the two crystals would need to be identical since otherwise, a grain boundary would form and coalescence would not occur. It happens that in 1989, three of us [13] observed and recorded the coalescence of identically oriented ^3He crystals without the large magnets which would be necessary for levitation. In this article, we present an analysis of these 1989 recordings. They concern several crystals with different radii at various temperatures close to 0.32 K. We have found excellent agreement with Maris' predictions [8].

Figure 1 shows the experimental cell used by Graner *et al.* [13] and Fig. 2 shows how a ^3He crystal was growing in it. At first sight, the growth looked like the flow of a viscous liquid, but it was not. The ^3He cell had two parts connected by a vertical capillary (see Fig. 1), and it was immersed in a ^4He liquid bath which provided a good thermal homogeneity. When increasing the pressure of liquid ^4He , the double membrane in the upper part of the ^3He cell was deformed and the ^3He pressure increased. When the ^3He crystallization pressure was reached, the first crystal seed happened to nucleate in the upper part. It was grown to a large size by increasing the ^4He pressure further. When the ^3He crystal was large enough, it started invading the lower part, and became visible at the lower

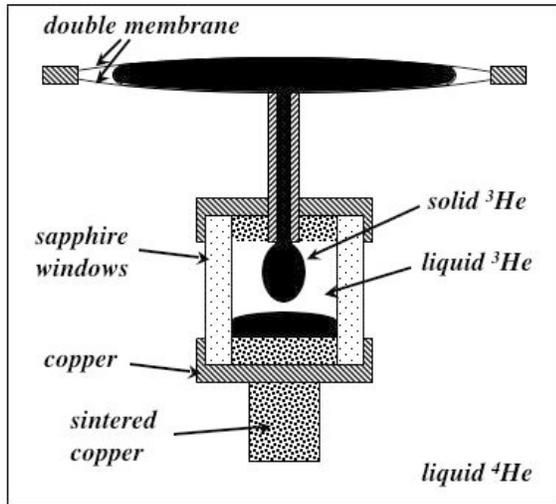


FIG. 1. The experimental cell where ^3He crystals were grown thanks to a flexible membrane. The lower part was 3.45 mm wide and 3.27 mm high; it could be observed through windows from the outside of the cryostat, as shown in Fig. 2.

end of the capillary [see Fig. 2(a)]. At that moment, the pressure was fixed and the shape evolved at constant *total* crystal volume. By exchanging mass with its liquid phase, i.e., by local growth and melting, the ^3He crystal started to move down because of gravity. The growth of usual crystals is driven by temperature gradients, but here it is different. The surrounding superfluid bath provides a good thermal homogeneity; furthermore, near the melting curve minimum, the liquid-solid transition only depends on pressure in a first order approximation (more precisely, the difference in chemical potential $\Delta\mu$ has no linear term in T). This explains why the shape evolution is driven only by gravity and surface tension.

At the surface of ^3He crystals, facets appear only below about 100 mK [14–19], so that their shape is rounded in all directions at 320 mK. They look like transparent honey flowing slowly down, although they are high quality single crystals. During their shape evolution, there is no plastic deformation of their lattice, only growth somewhere and melting somewhere else. It is precisely this kind of remarkable property that allowed unique studies of crystal surfaces in helium: equilibrium crystal shapes could often be reached very quickly (especially with ^4He crystals) [19]. These equilibrium shapes minimize the sum of gravitational and surface energies, exactly as for liquid drops.

For the “flow” of solid ^3He to proceed through the vertical capillary, a counter flow of mass was necessary because the liquid density is smaller. A liquid channel along the capillary wall could provide it. Another possibility is that vacancies move up inside the ^3He crystal, because their density and their mobility are large near T_{\min} [20]. After the crystalline drop reached the bottom of the cell (b), it divided into two crystals (c). This

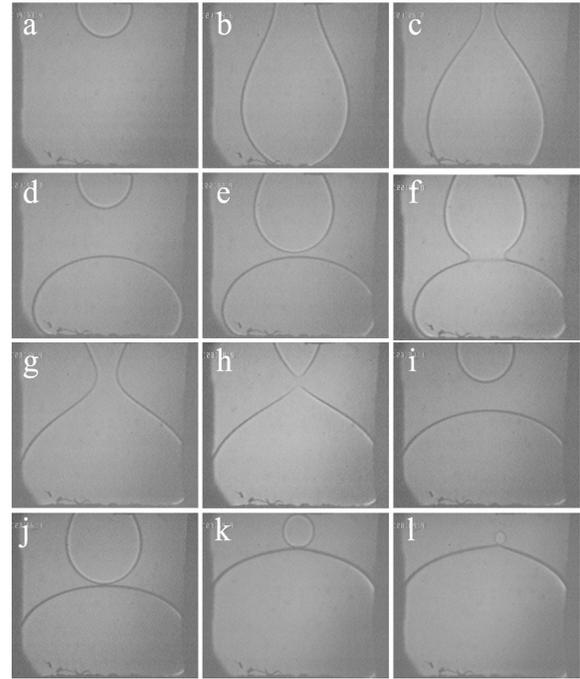


FIG. 2. Twelve images showing how a ^3He crystal “flows” down from the upper part of the cell into its lower part. For this recording, which took a few minutes, the temperature was $T_{\min} + 11$ mK. The crystal first “drips” down, so that a crystalline “drop” forms at the bottom (a)–(c); then a second drop appears (d) and comes into contact with the first one (e); coalescence is observed (f) and can be quantitatively analyzed (see Figs. 3 and 4). The second drop has exactly the same crystalline orientation as the first one because this is not real flow; it is local melting and growth of a single crystal which keeps the same orientation all the time. A third drop forms and coalesces again (g)–(j). Usually, the last drop (k) behaves differently because, being smaller than the orifice, it falls in the liquid and changes orientation before touching the lower crystal. As a consequence, there is a grain boundary between the two crystals which does not coalesce; the last drop keeps round, moves to the right, and finally vanishes (l). At this temperature, ^3He crystals have no facets; they are rough in all directions.

“dripping” must be due to the Rayleigh-Taylor instability, i.e., to the crystal neck being too narrow to be stable against capillary forces; we will analyze it in another article, because the shape is clearly different from what is observed with fluids and it deserves a detailed analysis. For the understanding of the present article, it is only important to realize that the lower crystal had exactly the same orientation as the upper one. The next picture in Fig. 2(d) shows that the lower crystal relaxed to an equilibrium shape at the bottom of the cell while the upper crystal kept “flowing” down until it came into contact with the lower one [Fig. 2(e)]. At this moment, there was no barrier against forming one single crystal again, since there was no grain boundary; the two crystals were nothing but two parts of the same previous crystal;

they had the same orientation. This is why we observed a rare and unusual event: the coalescence of two single crystals in a short time (Fig. 3).

After one coalescence event took place, there was some crystal remaining in the upper part of the cell, and it started to flow down again, so that another coalescence occurred [Fig. 2(g), 2(h), and 2(j)]. Usually, the last crystalline drops behaved differently [Fig. 2(k) and 2(l)]. They were smaller than the capillary diameter and fell down. During their motion they probably rotated slightly, so that when touching the lower crystal, a grain boundary formed and coalescence did not occur [Fig. 2(k)]. On the contrary, a cusp appeared between the two crystals and it took a much longer time for the small crystal to melt in favor of the large one [Fig. 2(l)]. If

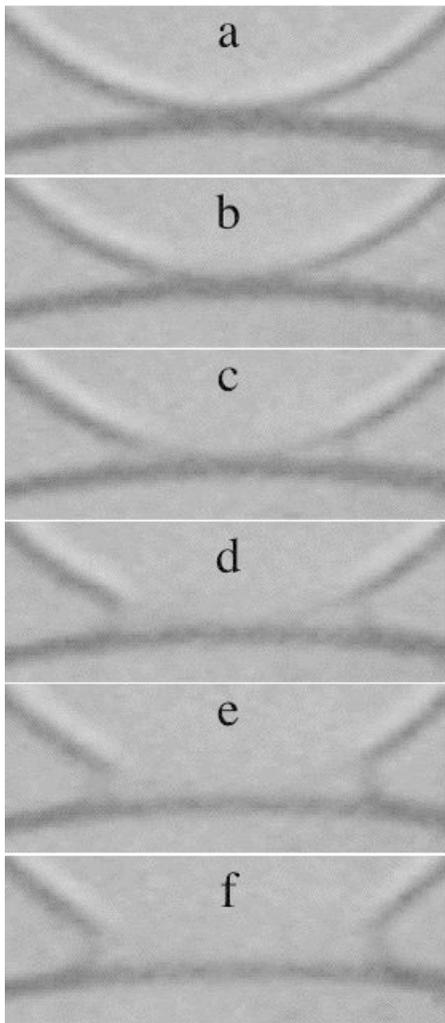


FIG. 3. Six successive images showing two ^3He crystals approaching each other (a), touching (b), and forming a neck which grows in time (c), (d), (e), and (f). The temperature is $T_{\min} + 11$ mK as for Fig. 2. Each image shows a region which is 0.2 mm by 0.5 mm in size. The time interval between two successive images is 20 ms.

needed, this proved that we were not dealing with viscous liquid matter but with single crystals. Note that, if we tried to do the same experiment with ^4He crystals, which are hcp instead of bcc, coalescence would not necessarily occur because stacking faults could form at the interface between two hcp crystals.

For an ordinary camera, the video rate is 50 fields per second, and coalescence takes typically 0.3 s; we could analyze ten events, each of them with about 15 images. Figure 3 shows six successive images. The two crystals approach [Fig. 3(a)], come into contact [Fig. 3(b)], and start coalescing [Fig. 3(c)–3(f)]. We have measured the neck radius from the outside of the black line which is due to refraction at the interface. For each event, we needed to determine the exact contact time $t = 0$ with an accuracy better than 20 ms. To do this, we tried various plots of all our data. R_n^3 versus time always gave well-defined linear behaviors at the beginning of the coalescence and a contact time equal to that of the last frame without visible neck ± 10 ms. This makes sense, in good agreement with the prediction by Maris [8]. We used these plots to determine the time $t = 0$ for each event. On the contrary, plots of R_n^2 versus time always gave infinite initial slope so that we could rule out a $t^{1/2}$ behavior of the neck radius. We also ruled out the $t \ln(\alpha t)$ behavior, because fits with this type of function gave contact times which were earlier by one to five frames than the last one without visible neck.

Given this qualitative agreement with Maris' predictions, we have proceeded with a quantitative check of Eq. (1). The coalescence is due to the growth of the neck at a velocity dR_n/dt under a difference in chemical potential which is proportional to the curvature of the solid-liquid surface. Note that gravity g plays a role only at scales larger than the capillary length $l_c = \sqrt{\gamma/\Delta\rho g} \approx 1$ mm, so that it can be neglected in the coalescence analysis. Furthermore, the surface tension is nearly isotropic, so that the neck is circular. At the beginning of the coalescence, the curvature of the interface is dominated by the curvature $1/r$ in the vertical plane. This is because the shape of the two crystals evolve slowly, except at the neck, where one curvature radius is $r \approx 2R_n^2/R$ while the other curvature radius is R_n , much larger at small time. The $t^{1/3}$ behavior is the consequence of dR_n/dt being proportional to $1/r$, i.e., to $1/R_n^2$. In our case, the two crystal radii R_1 and R_2 are different from each other, so that Eq. (1) has to be slightly modified. According to the above reasoning, one should simply replace the curvature $\kappa = 2/R$ in Eq. (1) by its more general form $(1/R_1 + 1/R_2)$.

Figure 4 shows a comparison of our experimental results with Maris' theory at four different temperatures, respectively $T - T_{\min} = -7$ mK, +11 mK, +16 mK, and +20 mK. Since Eq. (1) is only valid when the neck radius is much smaller than the crystal radii, we only kept data at small times ($t < 120$ ms). At larger times, the radius

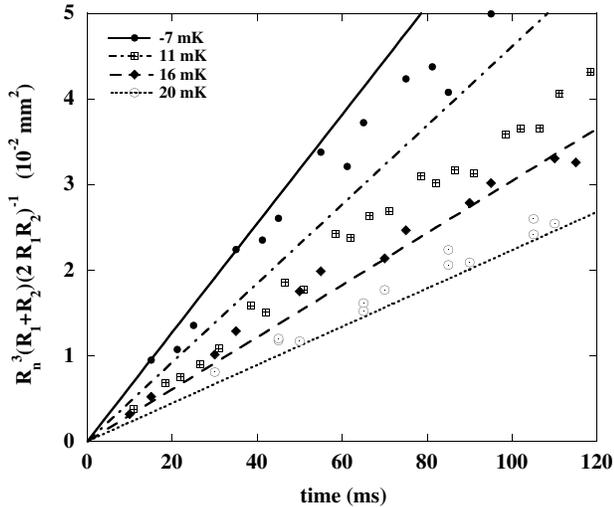


FIG. 4. The time variation of the cube of the neck radius R_n , normalized by the mean curvature radius $2R_1R_2/(R_1 + R_2)$ of the initial drops. Symbols correspond to different temperatures, respectively $T - T_{\min} = -7$ mK, $+11$ mK, $+16$ mK, and $+20$ mK with $T_{\min} = 320$ mK. Each group of data corresponds to two or three events where crystals had different radii. For each temperature, a line is also shown which corresponds to the quantity $3\pi k\gamma/\rho_C$, i.e., Maris' theory which has no adjustable parameter.

starts saturating, a phenomenon which could be anticipated from the weak curvature to the data in Fig. 4.

The coalescence is slower as the temperature difference $T - T_{\min}$ increases. This is because the growth coefficient k increases with the latent heat of crystallization; we used its experimental determination by Graner *et al.* [11–13]

$$k^{-1} = 550 + (3.9 \times 10^6)(T - T_{\min})^2, \quad (2)$$

with k^{-1} in cm s^{-1} and T in K. The experimental data represent ten recordings of coalescence with different initial radii of the drops. After normalization of R_n^3 by the mean curvature radius $2R_1R_2/(R_1 + R_2)$, we found consistency between all measurements. As can be seen, at each temperature, data corresponding to different initial radii agree not only with each other but also with theory. This agreement involves no adjustable parameter. Most of the experimental uncertainty comes from the temperature measurement, and its typical magnitude is given by the scatter in the data points.

In this article, we have presented experimental evidence that the time variation of the coalescence between two crystalline drops follows a critical behavior which is different from what one expects for viscous liquid drops. The coalescence is governed by surface curvature and surface mobility instead of viscous dissipation in the bulk or inertia. Our results show quantitative agreement with the $t^{1/3}$ prediction by Maris [8]. We believe that the same

critical behavior should occur with superfluid liquid drops, but coalescence would be so fast that the experiment would be rather difficult to perform. In a future article, we will describe dripping in the same system, which we have also observed to be quite different from the case of an ordinary liquid flow.

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