Analysis of the early stage of coalescence of helium drops

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We analyze the growth of the neck that forms between two liquid drops that have come into contact. The analysis is for a fluid in which the velocity of each point on the surface is proportional to the local curvature and directed normal to the interface. For this system, we show that the radius of the neck is proportional to $t^{1/3}$, where the time t is measured from the moment at which coalescence commences. We are able to find a simple expression for the shape of the interface in the vicinity of the neck.

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I. INTRODUCTION

When two liquid drops come into contact, a neck forms between them and grows rapidly. At later times, the fluid may remain as one connected mass or may break up [1]. The coalescence process is affected by the size of the drops, their relative velocity before impact, and their material properties, such as viscosity, density, and surface tension. In this paper, we are primarily interested in the very early stage of the coalescence process and, more specifically, in the variation of the radius R_n of the neck with the time t since coalescence of the drops began. For simplicity, we restrict attention to situations in which the impact velocity is zero and both drops have the same material properties and initial radius R.

Even then, there are a number of different cases that can be considered. If the drops are composed of classical fluids obeying the Navier-Stokes equation, it is necessary to solve this equation in the interior of the drop and with appropriate boundary conditions on the pressure and velocity fields at the drop surface. When the viscosity is sufficiently large, the fluid will move in a way such that at each instant the viscous forces very nearly balance the surface tension forces (Stokes' flow). In this regime, the density of the liquid does not affect the motion and so from dimensional analysis the neck radius R_n must vary as

$$R_n = R f(\sigma t/\eta R), \tag{1}$$

where σ is the surface tension, η is the viscosity, and f is a function whose form is to be determined. For a two-dimensional system, i.e., two long, parallel, fluid cylinders, it was shown by Hopper [2] that the function f has the form

$$f(x) = Ax|\ln(x)|, \tag{2}$$

where A is a constant. More recently, Eggers, Lister, and Stone [3] have shown that this result should also hold in three dimensions. Experiments to test the predictions of Eqs. (1) and (2) are under way [4]. Eggers *et al*. [3] point out that, regardless of the value of the viscosity, there will be Stokes' flow at very early times. At later times, if the liquid is not too viscous, there will be a transition to a regime in which the neck radius varies as $t^{1/2}$. Menchaca-Rocha *et al*. [5] studied the growth of the neck between two mercury drops and found results that are consistent with a $t^{1/2}$ law.

One can also consider the coalescence of two drops of superfluid helium. Depending on the temperature, superfluid helium is described by different hydrodynamic equations. There are at least three regimes to consider [6,7].

(1) At temperatures greater than 1 K, the motion of the liquid is well described by Landau's two-fluid model. These fluids, the so-called normal and superfluid components, have independent velocities $\vec{v}_n$ and $\vec{v}_s$, respectively. They occupy the same region of space and are able to pass through each other. The superfluid has no viscosity and has a velocity field $\vec{v}_s$ satisfying curl$\vec{v}_s = 0$. The normal fluid has a viscosity η_n. The normal and superfluid components satisfy coupled boundary conditions at the surface of the liquid. In this temperature range, a liquid drop is surrounded by a significant amount of helium vapor.

(2) At lower temperatures, the two-fluid model breaks down [6,7]. The normal fluid component is composed of elementary excitations (phonons and rotons). The excitations that make up the normal fluid have a well-defined collective velocity (normal fluid velocity) only when the number density of these excitations is sufficiently high that they make frequent collisions with each other. When the temperature is lowered, the mean free path of the elementary excitations (phonons and rotons) increases and can become larger than the dimensions of the drop. Under these conditions, the effect of the excitations is to exert a drag force on a moving surface that is normal to the surface and proportional to the surface velocity.

(3) Finally, at sufficiently low temperatures the effect of the excitations must become completely negligible.

The model we consider is a special case of the regime 2 of superfluid helium. We suppose that the liquid moves sufficiently slowly that there is always a balance between the surface tension force driving the motion and the drag force exerted by the excitations. Thus, this is analogous to the Stokes's flow of a classical fluid. Under these conditions, the velocity of the interface v_I will be in the direction outward normal to the interface and given by

$$v_I = \tilde{K}(\sigma\kappa + P), \tag{3}$$

where κ is the total curvature, taken as positive when the surface of the liquid is concave outward. We will call the constant $\tilde{K}$ the mobility of the interface. P is the pressure

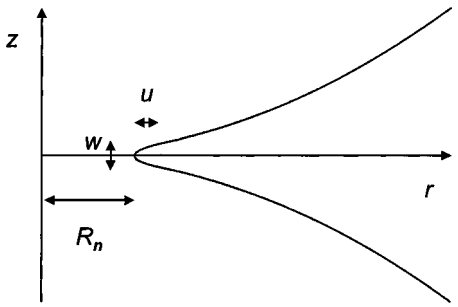

FIG. 1. Parameters describing the shape of the waist.

inside the drop, taken to be independent of position. We treat the liquid as incompressible so that P varies with time in such a way that the volume of the drop remains constant.

We note that, as well as being relevant to liquid helium, the model may be applicable to the motion of the interface between liquid and solid helium [8]. We discuss this in more detail in Sec. IV. In the mathematical literature the flow described by Eq. (3) is called "mean curvature flow" [9].

II. RATE OF GROWTH OF THE NECK

In the early stages of coalescence, the curvature in the vicinity of the neck will be much larger than the curvature of the remainder of the liquid surface. In addition, even though the neck is growing rapidly the change in volume of the liquid in the vicinity of the neck is very small compared to the total volume of the drops. Thus, the pressure remains nearly constant at the value $2\sigma/R$. Thus, we can make the approximation that the only part of the surface that is moving is the part near the neck. It follows that, when the neck has a radius R_n, the width w of the neck will be (for definition, see Fig. 1)

$$w = R_n^2/R. \tag{4}$$

The surface tension force pulling outward on a section of the circumference of the neck of length δC will be $2\sigma\delta C$. This has to be balanced by the drag force acting on the neck. This drag force is proportional to the area of the neck, $w\delta C$, and to the rate of growth of the neck, and inversely proportional to $\tilde{K}$. Thus,

$$2\sigma\delta C = a\frac{w\delta C}{\tilde{K}}\frac{dR_n}{dt}, \tag{5}$$

where a is a dimensionless coefficient dependent on the shape of the neck. Let us assume for the moment that a is independent of the neck radius and hence also independent of the time since coalescence began. Then, combining Eqs. (4) and (5), we have

$$R_n^2\frac{dR_n}{dt} = \frac{2\tilde{K}\sigma R}{a}, \tag{6}$$

and so

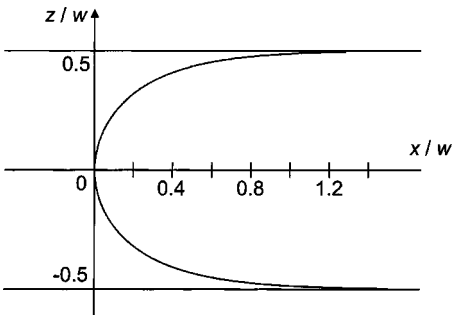

FIG. 2. Coordinate system used in the calculation of the shape of a surface moving down a channel with parallel walls separated by a distance w. The shape of the interface is as given by Eq. (14).

$$R_n = \left(\frac{6\tilde{K}\sigma R t}{a}\right)^{1/3}. \tag{7}$$

Now consider the assumption that the coefficient a remains constant. This will hold true if the shape of the neck remains constant as the radius of the neck grows. Thus, for example, with reference to Fig. 1, does the "depth" of the neck u vary with time in the same way as does w? To answer this question, we note that, if w were held constant, the time t_n for the neck to come to an equilibrium shape would be of the order of w divided by the surface velocity. Since the surface velocity is of the order of $\tilde{K}\sigma/w$, we have

$$t_n \sim w^2/\tilde{K}\sigma \sim \frac{R_n^4}{R^2\tilde{K}\sigma}. \tag{8}$$

This is to be compared to the time scale t_w on which the width of the neck changes, which is given by

$$t_w = \frac{w}{dw/dt} = \frac{R_n}{2dR_n/dt} = \frac{R_n^3 a}{4R\tilde{K}\sigma}. \tag{9}$$

It can be seen that t_n is smaller than t_w by a factor of R_n/R. Thus in the early stages of coalescence it appears from this argument that the shape of the neck should be constant.

We now determine the coefficient a. Based on the above arguments, it should be sufficient to do this for a surface advancing down a channel of constant width w with the liquid having zero contact angle with the walls. Let x be the distance along the channel and z across, with the middle of the channel at $z=0$ (Fig. 2). Suppose that the interface is moving along the channel with a constant velocity v. Let the velocity of the interface in the direction normal to the surface be v_I and let the angle that the tangent to the interface makes to the x axis be θ. Then

$$v_I = v\sin\theta = \tilde{K}\sigma\kappa. \tag{10}$$

We have $\sin\theta = (dz/dx)/[1+(dz/dx)^2]^{1/2}$ and the curvature of the surface is $\kappa = -(d^2z/dx^2)/[1+(dz/dx)^2]^{3/2}$. Hence, from Eq. (3) (and setting $P=0$ for the moment)

$$\frac{d^2z}{dx^2}\frac{dx}{dz}\frac{1}{1+(dz/dx)^2}=-\frac{v}{\tilde{K}\sigma}. \tag{11}$$

The right-hand side of this equation is a constant, and this equation provides a differential equation that determines the shape of the moving surface. To solve Eq. (11), set $dz/dx=f$, so the equation becomes

$$\frac{df}{dx}=-\frac{v}{\tilde{K}\sigma}f(1+f^2). \tag{12}$$

Then, by integration we get

$$f=\pm\frac{1}{[\exp(2vx/\tilde{K}\sigma)-1]^{1/2}}, \tag{13}$$

where we have chosen the constant of integration so that $|dz/dx|=\infty$ at $x=0$. A second integration gives

$$z=\pm\frac{\tilde{K}\sigma}{v}\tan^{-1}\{[\exp(2\pi x/w)-1]^{1/2}\}. \tag{14}$$

As $x\to\infty$, $z\to w/2$. Therefore the velocity v must equal $\pi\tilde{K}\sigma/w$ and so the shape of the surface is

$$z=\pm\frac{w}{\pi}\tan^{-1}\{[\exp(2\pi x/w)-1]^{1/2}\}. \tag{15}$$

This is the interface shape shown in Fig. 2.

From a comparison of the above analysis with Eq. (5), we obtain

$$a=\frac{2}{\pi}, \tag{16}$$

and so the final result for the radius of the neck is

$$R_n=(3\pi\tilde{K}\sigma Rt)^{1/3}. \tag{17}$$

The shape of the neck for the coalescing drops can then be written in cylindrical polar coordinates (r,z) by using Eq. (4) for w, i.e., by setting $w=R_n^2/R$, to obtain

$$z=\pm\frac{R_n^2}{\pi R}\tan^{-1}(\{[\exp[2\pi(r-R_n)R/R_n^2]-1\}^{1/2}). \tag{18}$$

This result is based on setting the pressure equal to zero in Eq. (3). As mentioned earlier, the pressure at any instant must adjust to a value that ensures that the volume of the drop remains constant. A change in the pressure will result in a motion of the entire surface of the drop, not just in the region near to the neck. The pressure must be negative so that the main part of the drop contracts to compensate for the increase ΔV in the volume in the vicinity of the neck. From Eq. (4), we have to lowest order in R_n/R

$$\Delta V=\frac{\pi R_n^4}{2R}. \tag{19}$$

Hence in order for the total volume to remain constant, the radius of each drop must shrink by an amount

$$\Delta R=-\frac{R_n^4}{16R^3}. \tag{20}$$

This is smaller than the width w of the waist by a factor of the order of R_n^2/R^2. Thus it appears that at early time the effect of the pressure can be neglected.

Finally, we note that, since we are considering only what happens in the vicinity of the neck, we could as an alternative set $w=r^2/R$ in Eq. (15), to obtain the formula

$$z=\pm\frac{r^2}{\pi R}\tan^{-1}(\{[\exp[2\pi(r-R_n)R/r^2]-1]\}^{1/2}). \tag{21}$$

This formula has the feature that when the argument of the inverse tangent becomes large, i.e., when r lies outside the neck region, the relation between z and r becomes

$$z\approx\pm\frac{r^2}{2R}. \tag{22}$$

Thus the solution smoothly goes over to the equation for the undisturbed surface of the drops near to the point of contact.

III. NUMERICAL SIMULATION

We have performed a computer simulation to compare with the analytical calculation given above. To avoid having to deal with a singular initial shape, we started with a neck of radius R_{n0} and took the starting position of the surface to be given by the equations

$$r=\begin{cases} R_{n0}+bz^2 & |z|<z_c\,, \quad (23)\\ (2|z|R-z^2)^{1/2} & |z|>z_c\,. \quad (24)\end{cases}$$

By choosing

$$b=\frac{1}{2z_c}\frac{R-z_c}{(2z_cR-z_c^2)^{1/2}} \tag{25}$$

and

$$R_{n0}=(2z_cR-z_c^2)^{1/2}-bz_c^2, \tag{26}$$

it follows that Eqs. (23) and (24) give a curve that is continuous at z_c and which also has a continuous derivative at this point. The starting radius of the waist was $0.01R$. In performing the simulation it was assumed that the drops retained axial symmetry as coalescence progressed and that $r(-z)=r(z)$. Thus, in both the simulation and the analytical calculation, it is implicitly assumed that there are no instabilities that break these symmetries. A set of 7800 points on the surface was used. The result for the radius of the neck as a function of time is shown in Fig. 3, along with the analytical result Eq. (16). It can be seen that the agreement when the waist is small is excellent. In the range where the waist is above about $0.06R$, the waist obtained from the numerical simulation is smaller than that predicted by the analytical result. This is to be expected since the analytical result is

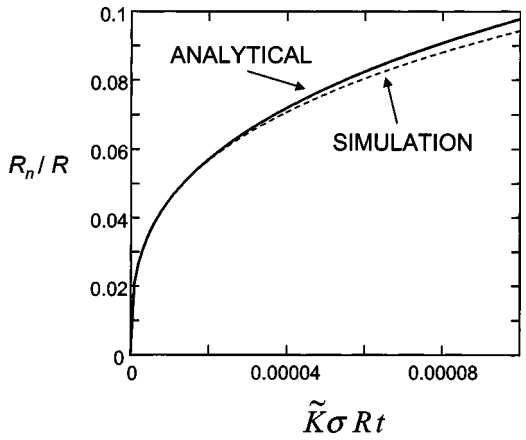

FIG. 3. Radius of the waist R_n divided by initial radius of the drop R as a function of time t multiplied by $\tilde{K}\sigma R$. The dashed curve shows the result from the computer simulation and the solid curve is the analytical result [Eq. (16)].

valid only when the waist radius is much less than R. Equation (17), which predicts a waist radius increasing for all times, must give too large a value of R_n for large t.

In Fig. 4 we show a comparison of the shape of the neck region obtained from the numerical simulation and from the analytical calculations [Eqs. (18) and (21)]. The comparison is made when $\tilde{K}\sigma Rt=0.000\,01$ at which time the waist radius is $R_n=0.0454R$. The agreement is excellent. Based on the analytic solution, the radius should increase as $t^{1/3}$ and the width of the neck should vary as $t^{2/3}$. Thus the shape of the neck expressed in terms of the reduced variables $r/t^{1/3}$ and $z/t^{2/3}$ should be independent of time for small times. A plot of this type is shown in Fig. 5. It can be seen from this figure that the shape of the neck in these reduced variables is indeed almost independent of time. Note that for the largest time for which $\tilde{K}\sigma Rt=0.0001$, the radius R_n of the neck is $\sim 0.1R$ and is increasing with time slightly less rapidly than as $t^{1/3}$. Thus, when the shape of the neck is plotted in the reduced variables, the neck radius shrinks slightly as time

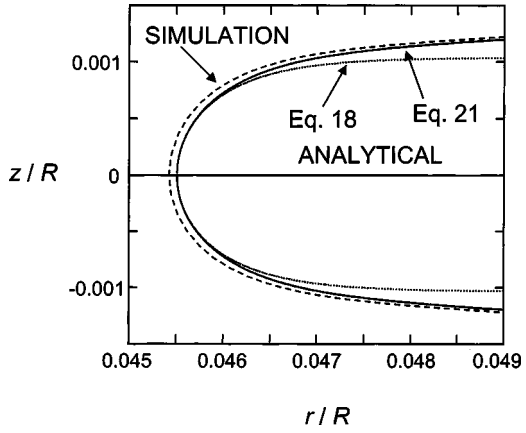

FIG. 4. Shape of the region around the waist at a time such that $\tilde{K}\sigma Rt=0.000\,01$. The dashed curve shows the result from the computer simulation and the analytical results from Eqs. (18) and (21) are shown by the dotted and solid curves, respectively.

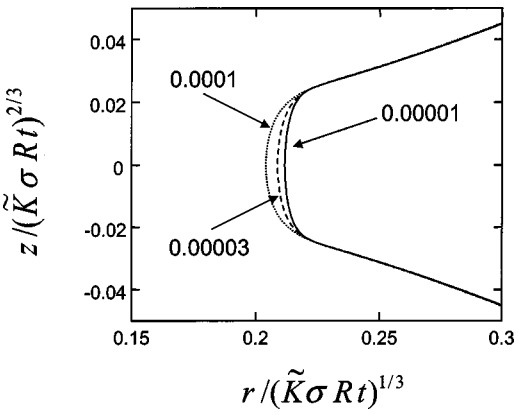

FIG. 5. The shape of the neck expressed in terms of the reduced variables $r/(\tilde{K}\sigma Rt)^{1/3}$ and $z/(\tilde{K}\sigma Rt)^{2/3}$ for times such that $\tilde{K}\sigma Rt=0.000\,01$, $0.000\,03$, and 0.0001.

increases. Finally, in Fig. 6 we show the shape of the drops at much later times.

IV. POSSIBLE APPLICATION TO THE LIQUID-SOLID HELIUM INTERFACE

The interface between liquid and solid helium has a number of unique properties and has been studied in great detail. Helium is the only substance that can remain liquid down to absolute zero temperature. For ^{4}He at zero temperature, it is necessary to apply a pressure of 25 bars in order to form the solid. By introducing an appropriate amount of helium into a cell of fixed volume, it is possible to have liquid and solid coexisting. Under normal gravitational conditions, the solid will occupy the bottom of the experimental cell because of its higher density. However, it should be possible to levitate drops of the solid phase optically or magnetically as has been done for the liquid [10,11], and to study the coalescence of these drops. Of course, in considering the coalescence of solid drops, it is necessary to allow for the variation of the surface energy over the surface of the drop.

As discussed by Castaing and Nozieres [12], the growth rate of solid from the liquid is determined by the difference $\Delta\mu$ in the chemical potential per unit mass of the two phases and the temperature difference ΔT. When these differences are sufficiently small,

$$\frac{\Delta\mu}{T}=aJ+bJ_E\,, \tag{27}$$

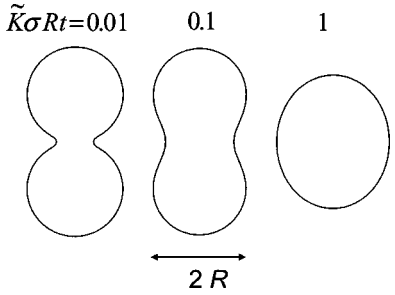

FIG. 6. The shape of the drops at times such that $\tilde{K}\sigma Rt$ has the values 0.01, 0.1, and 1. The initial diameter of each drop is $2R$.

$$\frac{\Delta T}{T^2} = bJ + cJ_E , \tag{28}$$

where J and J_E are the currents of mass and entropy, respectively, flowing across the interface, and a, b, and c are Onsager coefficients. Note that the same coefficient b appears in both equations. We ignore the thermal effects for the moment so that we can write the velocity v of the interface as

$$v = K\Delta\mu . \tag{29}$$

In the literature on solid helium, K is referred to as the growth coefficient [13], and is related to the coefficient $\tilde{K}$ previously introduced by

$$\frac{K}{\rho_s} = \tilde{K} . \tag{30}$$

For a classical substance, the growth rate is determined by the rate at which atoms in the liquid can move to find correct positions on the solid surface. Thus, K increases with increasing temperature. For helium, on the other hand, one can consider that freezing amounts to the smooth conversion of one quantum state (the liquid) into another (the solid), and so at zero temperature there is no dissipation associated with this process. For $T \neq 0$, a finite growth coefficient arises because as the interface moves thermal excitations coming from either the liquid or solid side are reflected and exert a drag force on the interface. In the solid these excitations are phonons, and in the liquid they are phonons and rotons [14,15].

Now we consider the conditions under which the motion of the interface will be sufficiently slow that at each moment the surface tension forces nearly balance the drag force on the interface. It is convenient to do this by analyzing the small amplitude motion of a planar interface. From the work of Andreev and Parshin [14], the dispersion relation for these so-called ''melting-freezing waves'' is obtained from the solution of the equation

$$\omega^2 + \frac{i\omega k\rho_s\rho_l}{K(\rho_s-\rho_l)^2} - \frac{k^3\sigma\rho_l}{(\rho_s-\rho_l)^2} = 0, \tag{31}$$

where ω is the frequency, k is the wave number, σ is now the liquid-solid surface energy, and ρ_s and ρ_l are the densities of the solid and liquid, respectively. Let

$$k_c \equiv \frac{\rho_s^2\rho_l}{4\sigma K^2(\rho_s-\rho_l)^2} . \tag{32}$$

Then for $k > k_c$ Eq. (28) has a pair of solutions of the form $\omega = \pm\omega_R + i\omega_I$, where ω_I is negative. These solutions correspond to damped propagating waves traveling along the interface. For $k < k_c$, the solutions for ω are purely imaginary, and for $k \ll k_c$ a disturbance of the interface with wave number k relaxes with a frequency

$$\omega = -\frac{ik^2K\sigma}{\rho_l} = -ik^2\tilde{K}\sigma . \tag{33}$$

Thus, in this range of wave numbers the interface moves at a velocity which is proportional to the local curvature, and the theory that we have developed should be applicable. Hence, from this analysis we conclude that during the coalescence process the features of the interface that can be described with $k < k_c$ will be adequately described by our theory, whereas very sharp features with $k > k_c$ will not. For ^{4}He at 1.35 K, for example, $K^{-1} \approx 10^3$ cm s^{-1} [16], and we obtain $k_c \approx 3\times 10^7$ cm^{-1}. Thus, our theory should give a good description apart from details on a length scale of a few angstroms, which are unlikely to be observable in any case. As the temperature is lowered, K increases, k_c decreases, and eventually k_c^{-1} becomes comparable to the radius of the drop and the theory is completely inapplicable.

We can now consider the neglect of thermal effects, i.e., the replacement of Eqs. (27) and (28) by Eq. (29). This relies on the assumption that either the solid or the liquid (or preferably both) are good heat conductors so that the latent heat that is liberated when the solid is formed is efficiently carried away. We have not investigated this in detail. However, we note that for ^{3}He at around 0.32 K, the latent heat becomes zero [6] thus making heat conduction unnecessary. At this temperature [17] $K^{-1} \approx 500$ cm s^{-1} and so k_c is again very small, making the theory applicable. Of course, since ^{3}He is not superfluid at this temperature, it may be necessary to consider the effects of the viscosity of the liquid on the coalescence process.

The damping of the interface arises because elementary excitations (phonons and rotons) bounce off the surface and damp its motion. For this process, the drag force on the interface is proportional to the surface velocity only when the surface velocity is small compared to the propagation velocity $v_{\rm ex}$ of the excitations. Since the radius of the neck varies as $t^{1/3}$, it follows that within the model the surface velocity varies as $t^{-2/3}$. Thus, the results we have derived should be used only for times such that this velocity is much less than the excitation velocity, which means for times such that

$$t \gg \frac{(3\pi\tilde{K}\sigma R)^{1/2}}{(3v_{\rm ex})^{3/2}} , \tag{34}$$

or

$$\frac{R_n}{R} \gg \left(\frac{\pi K\sigma}{v_{\rm ex}\rho_s R}\right)^{1/2} . \tag{35}$$

For $K^{-1} = 10^3$ cm s^{-1}, a typical excitation velocity $v_{\rm ex}$ of 10^4 cm s^{-1}, $\sigma = 0.17$ erg cm^{-2}, and $R = 1$ cm, this means that it is necessary to have $R_n \gg 5\times 10^{-4}R$.

V. SUMMARY

We performed a calculation of the early stage of coalescence of two drops. The calculation is performed for a liquid whose interface moves at a velocity that is proportional to the local curvature. We are able to obtain an analytical solution valid when the radius of the neck is much less than the initial radius of the drops. The calculated shape of the neck

and rate of growth are in excellent agreement with the results from a numerical simulation. The theoretical predictions of this paper can be tested through studies of the coalescence of liquid or solid helium drops in an appropriate temperature range.

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[1] For experimental work, see, for example, J. Qian and C. K. Law, J. Fluid Mech. **331**, 59 (1997).
[2] R. W. Hopper, J. Fluid Mech. **213**, 349 (1990); **243**, 171 (1992); J. Am. Ceram. Soc. **76**, 2947 (1993); **76**, 2953 (1993).
[3] J. Eggers, J. R. Lister, and H. A. Stone, J. Fluid Mech. **401**, 293 (1999).
[4] W.-J. Yao, P. Pennington, H. J. Maris, and G. M. Seidel (unpublished).
[5] A. Menchaca-Rocha, A. Martinez-Davalos, R. Nunez, S. Popinet, and S. Zaleski, Phys. Rev. E **63**, 046309 (2001).
[6] J. Wilks, *The Properties of Liquid and Solid Helium* (Oxford University Press, London, 1967).
[7] I. M. Khalatnikov, *An Introduction to the Theory of Superfluidity* (Benjamin, New York, 1965).
[8] This statement ignores the fact that the both the surface energy and the coefficient K depend on the orientation of the surface with respect to the crystallographic axes.
[9] K. Ishii and H. M. Soner, SIAM J. Math. Anal. **30**, 19 (1998).
[10] M. A. Weilert, D. L. Whitaker, H. J. Maris, and G. M. Seidel, J. Low Temp. Phys. **98**, 17 (1995).
[11] M. A. Weilert, D. L. Whitaker, H. J. Maris, and G. M. Seidel, J. Low Temp. Phys. **106**, 101 (1997); Phys. Rev. Lett. **77**, 4840 (1996).
[12] B. Castaing and P. Nozieres, J. Phys. (France) **41**, 701 (1980).
[13] Note that, in some papers, μ is the chemical potential per unit mass and in others the potential per atom.
[14] A. F. Andreev and A. Y. Parshin, Sov. Phys. JETP **48**, 763 (1978).
[15] A. Y. Parshin, Physica B & C **109&110B**, 1819 (1982).
[16] J. Bodensohn, P. Leiderer, and D. Savignac, in *Proceedings of the 4th International Conference on Phonon Scattering in Condensed Matter*, edited by W. Eisenmenger, K. Lassmann, and S. Dottinger (Springer, Berlin, 1984), p. 266.
[17] F. Graner, S. Balibar, and E. Rolley, J. Low Temp. Phys. **75**, 69 (1989); S. Balibar, D. O. Edwards, F. Graner, and E. Rolley, in *Excitations in Two-Dimensional and Three-Dimensional Quantum Fluids*, edited by A. F. G. Wyatt and H. J. Lauter (Plenum, New York, 1991), p. 375; S. Balibar, D. O. Edwards, and W. F. Saam, J. Low Temp. Phys. **82**, 119 (1991).