Nucleation of Bubbles in Liquid Helium at Negative Pressure

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We consider the rate at which bubbles form when a negative pressure is applied to liquid ^{4}He. We show that at a critical pressure P_c of about -9 bars, the liquid becomes macroscopically unstable and the barrier against nucleation of bubbles becomes zero. The tensile strength of liquid helium is calculated as a function of temperature allowing for these effects.

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How large a negative pressure can be applied to liquid helium before bubbles form? This is determined by the rate at which bubbles nucleate, either over the nucleation barrier as a result of thermal fluctuations or via quantum tunneling. There has been considerable interest in this topic[1] because of the possibility that at low temperatures the nucleation rate, and hence the tensile strength, might be determined by the quantum tunneling rate. It has been predicted[2] that quantum tunneling should dominate at temperatures below 0.3 K, and that in this temperature range the tensile strength should be more than 15 bars, i.e., bubbles will not form until the pressure is less than -15 bars. In this Letter we show that before this pressure is reached ^{4}He becomes macroscopically unstable. We present a calculation of the nucleation rates and tensile strength allowing for this effect.

Currently, the best information about the equation of state of liquid helium at $T=0$ K comes from the measurements of the sound velocity c as a function of pressure P made by Abraham *et al.*[3] From these extremely accurate data one can find $P(\rho)$ and $E(\rho)$ (ρ is the density, E is the internal energy per unit mass) by standard thermodynamic relations.[3] To determine the equation of state of helium for negative pressure, we first extrapolate the $c(P)$ data for positive P into this region. One can summarize the results of attempts to do this as follows.[4] Simple polynomial fits do not lead to consistent estimates of $c(P)$ for $P \lesssim -2$ bars. Padé approximants do better and suggest that c goes to zero at around -10 bars (± 2 bars) with a fractional power law. At first sight this behavior is surprising, but actually it is to be expected even on the simplest model for the equation of state. The pressure vanishes at the equilibrium density of 0.145 13 g cm^{-3} and must also be zero at $\rho = 0$. Thus, at some density ρ_c between 0 and ρ_0 the pressure must have a maximum negative value P_c. If we assume for the moment that E is an analytical function of ρ we can use the relations $P = \rho^2 \partial E/\partial\rho$ and $c^2 = \partial P/\partial\rho$ to show that for ρ slightly larger than ρ_c

$$P - P_c \propto (\rho - \rho_c)^2, \quad (1)$$

$$c \propto (\rho - \rho_c)^{1/2}. \quad (2)$$

Consequently, the second velocity goes to zero as

$$c \propto (P - P_c)^{1/4}. \quad (3)$$

Thus, one should be able to make an extrapolation of c into the negative pressure range more reliably by making a fit of c^4 as a function of P. We have done this[4] using several methods (Padé and polynomials of various orders) and the results are reasonably independent of the method. In Fig. 1 we show a 2-2 Padé fit to the experimental data. P_c is found to be -8.9 bars, and we believe this is accurate to about ± 1 bar. The critical density is 0.11 g cm^{-3} and at this density the internal energy per particle is estimated to be -6.5 K per particle.

The vanishing of the sound velocity means that the liquid becomes macroscopically unstable at P_c, i.e., un-

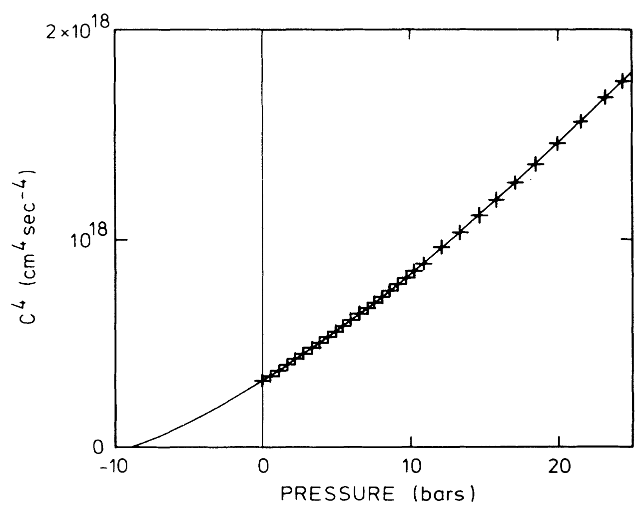

FIG. 1. Plot of c^4 (c = sound velocity) vs pressure. Data are from Ref. 3. Solid line is a Padé fit. Because of the large number of data points at low P not all points have been plotted.

stable at long wavelengths. Thus, it is clear that the predictions of previous theories that pressures as negative as −15 bars can be achieved must be incorrect. But the interesting question is how does the instability affect the nucleation process? In the "standard theory" one considers the energy F required to form a bubble of radius R. This is

$$F=4\pi R^2\alpha-\tfrac{4}{3}\pi R^3|P|\,, \tag{4}$$

where α is the surface energy. This has a maximum at $R=2\alpha/|P|$, and so the energy barrier is

$$\Delta F=16\pi\alpha^3/3|P|^2\,. \tag{5}$$

This barrier height does not involve the equation of state, and remains finite even for $|P|>|P_c|$. The mistake in the calculation is the implicit assumption that the most favorable way for nucleation to occur is through the formation of an "ideal bubble," i.e., a bubble with a wall of negligible thickness surrounding a region of zero density. In a more general approach one can consider the creation of a bubble in which the density is a smoothly varying function of the radius r. For the energy increase resulting from this density variation we take[5]

$$F=\int dV[\phi(\rho)+\lambda|\nabla\rho|^2]\,, \tag{6}$$

where $\phi(\rho)$ is the energy per unit volume for a system with uniform density ρ, and λ is a constant describing the energy associated with density gradients. We now vary ρ and look for the path that proceeds to a large bubble by passing over a barrier of minimum height.

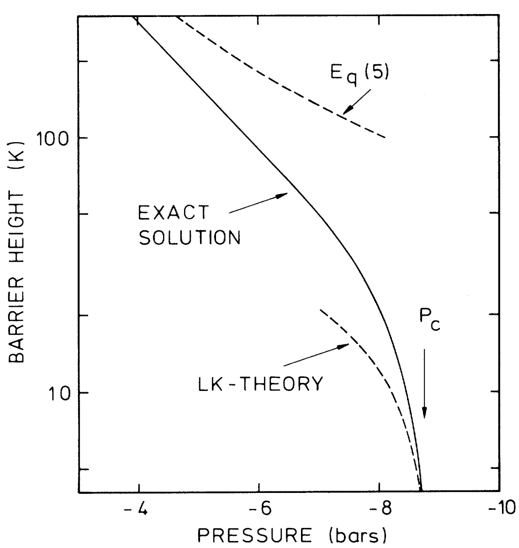

FIG. 2. Barrier height ΔF for nucleation of a bubble as a function of pressure. The dashed lines show the barrier calculated for a bubble with a thin wall [Eq. (5)], and that according to the Lifshitz-Kagan theory, valid near P_c. The solid line is the exact solution.

This height is then the barrier ΔF. It is straightforward to show that the density configuration corresponding to the critical nucleus of energy ΔF is the solution of the equation

$$\frac{1}{r^2}\frac{d}{dr}\left(r^2\frac{d\rho}{dr}\right)=\frac{1}{2\lambda}\frac{d\phi}{d\rho}\,. \tag{7}$$

Thus, to find ΔF one must solve (7) and substitute the solution into Eq. (6).

To do this we have determined[4] $\phi(\rho)$ from the extrapolated sound velocity in the region $\rho_0>\rho>\rho_c$. For $\rho<\rho_c$ we have approximated ϕ by a polynomial that fits smoothly onto ϕ for $\rho>\rho_c$. We then fixed λ by requiring that the energy of a planar free surface of the liquid have the correct value[6] (0.378 erg cm^{-2}). This gives $\lambda=9.13\times10^{-7}$ erg. The final results for the barrier height ΔF are shown in Fig. 2 as the solid line, together with the predictions of Eq. (5).

For small $|P|$ (e.g., 1 or 2 bars) the numerical results are in reasonable agreement with the barrier given by Eq. (5). The critical nucleus is large compared to the wall thickness of the planar interface. At larger $|P|$ the difference between the numerical solution and Eq. (5) increases, and as $P\rightarrow P_c$ the barrier tends to zero. In this range the critical nucleus has a density at its center which is only slightly less than the density in the bulk liquid. Also, the size of the nucleus no longer decreases as $|P|$ is raised, but actually increases slowly as $P\rightarrow P_c$. In the region P very close to P_c we can compare the numerical solution with an analytical calculation of Lifshitz and Kagan,[5] who derived a general result for the barrier valid when a system is close to an instability. We find agreement with their result, but only for P *extremely*

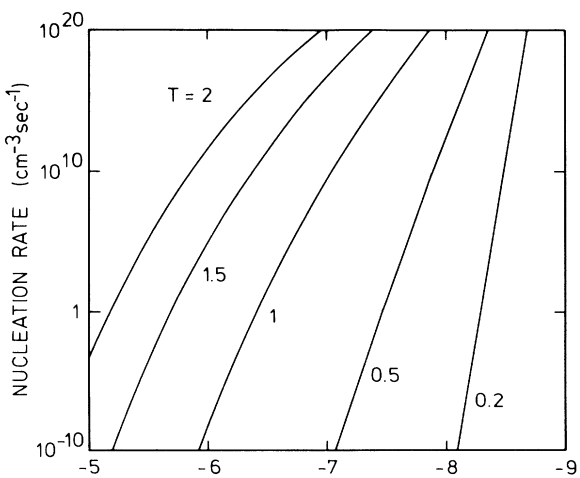

FIG. 3. Calculated nucleation rate per unit volume and time as a function of pressure. The different curves are labeled by the temperature.

close to P_c (see Fig. 2). In this regime the bubble size goes as $(P-P_c)^{-1/4}$.

From the energy barrier we can calculate the nucleation rate using

$$\Gamma = \Gamma_0 \exp(-\Delta F/kT) . \quad (8)$$

For simplicity we take the prefactor to be the product of an attempt frequency kT/h with a density of nuclei equal to the reciprocal of the volume of a sphere of radius 10 Å (roughly the bubble size). This gives the results shown in Fig. 3. To obtain the effective tensile strength one has to consider the experimental situation. Suppose a negative pressure is applied to a volume V of liquid for time τ. Nucleation of a bubble is likely to occur if $\Gamma V \tau$ is of the order of 1. Thus, the tensile strength is the magnitude of the pressure at which this occurs. We show this in Fig. 4 for $V\tau$ equal to 1 and 10^{-10} cm^{3}sec. Because Γ rises so rapidly with increasing $|P|$, the difference in the tensile strength for the two different values of $V\tau$ is fairly small. We include in Fig. 4 the tensile strength based on the ideal-bubble value for ΔF [Eq. (5)]. As already discussed, this gives a strength which increases very rapidly as T is lowered.

We have also considered[4] the effect of quantum tunneling through the barrier. In earlier theories[1,2] this was assumed to be the process that limited the rise in the tensile strength at low temperatures, whereas it is now clear that this rise is limited by the finite value of P_c. We estimate that the effect of quantum nucleation is limited to the small region of the P-T plane below ≈ 0.3 K and for P very close to P_c. Thus, it will be extremely hard to study this quantum nucleation since controlled and accurately known pressures near P_c are required.

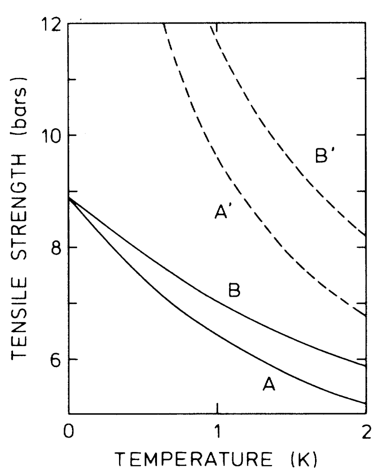

FIG. 4. Calculated tensile strength of liquid helium as a function of temperature. Solid lines are the results of the full calculation based upon Eqs. (6) and (7). Dashed lines show the tensile strength assuming that the critical nucleus is an ideal bubble [Eq. (5)]. A,A' and B,B' are for products of experimental time and volume of 1 and 10^{-10} cm^{3}sec, respectively.

The current experimental situation is as follows. Most of the early measurements of the tensile strength gave very low values. This has been attributed[1,2,7] to the use of large experimental volumes V that were likely to contain vortices or electron bubbles (produced by cosmic rays, for example). Recently, much better measurements have been made by Nissen *et al.*,[8] who used an ultrasonic technique to study a volume of roughly 10^{-5} cm^3 for times of the order of 10^{-6} sec. They obtained tensile strengths in the range 6 to 8 bars for temperatures decreasing from 2.2 down to 1.6 K. They estimated that the error in determination of the strength was less than 20%, but the uncertainty may actually be larger than this because they have not allowed for the peculiarities of the equation of state for negative pressures that we have discussed here.[9] In the temperature range 1.6 to 2.2 K the difference between the theory presented here and the earlier calculations is not large enough for the experimental data to discriminate between the two theories (see Fig. 4). However, it is clear that application of the same experimental technique at lower temperatures would allow a clear distinction to be made and provide a test of our estimate of P_c.

Finally, we mention some interesting related topics. In ^{3}He the sound-velocity data[10] are not as accurate, and our best estimate of P_c is -3 bars (± 1 bar). Both in ^{3}He and in ^{4}He the analysis[4] of the sound velocity suggests that c may go to zero as $(P-P_c)^\nu$ with ν slightly larger than $\frac{1}{4}$, around 0.30–0.32. This would indicate that near the instability point the energy may not be an analytical function of density, but we know of no theory of this for a quantum system at $T=0$ K.

In summary, we have shown that at a pressure of approximately -9 bars liquid ^{4}He becomes unstable at long wavelengths, and we have calculated how this affects the rate of nucleation of bubbles in the liquid. Measurements of the nucleation rate at low temperatures ($T \leq 1$ K) would test the theory and provide new insight into the equation of state of liquid helium.

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[1] For a review of cavitation in quantum liquids, see V. A. Akulichev, Ultrasonics **24**, 8 (1986).

[2] V. A. Akulichev and V. A. Bulanov, Akust. Zh. **20**, 817 (1975) [Sov. Phys. Acoust. **20**, 501 (1975)].

[3] B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and P. R. Roach, Phys. Rev. A **1**, 250 (1970).

[4] Q. Xiong and H. J. Maris (to be published).

[5] I. M. Lifshitz and Y. Kagan, Zh. Eksp. Teor. Fiz. **62**, 385

(1972) [Sov. Phys. JETP **35**, 206 (1972)].

[6]J. R. Eckardt, D. O. Edwards, S. Y. Shen, and F. M. Gasparini, Phys. Rev. B **16**, 1944 (1977). We have used the zero-temperature value of the surface energy throughout the calculations. In the same spirit we have neglected the temperature dependence of the equation of state.

[7]P. L. Martson, J. Low Temp. Phys. **25**, 383 (1976).

[8]J. A. Nissen, E. Bedegom, L. C. Brodie, and J. S. Semura, Adv. Cryog. Eng. **33**, 999 (1988).

[9]To determine the tensile strength it is necessary to estimate the size of the pressure fluctuation at the focus of the ultrasonic transducer. In Ref. 8 this was done by calculation of the focusing factor of the transducer and also by measuring the scattering of light from the focal region. Both of these methods require a knowledge of the equation of state for negative pressures.

[10]B. M. Abraham, D. Chung, Y. Eckstein, J. B. Ketterson, and P. R. Roach, J. Low Temp. Phys. **6**, 521 (1972).