

Experimental investigation of cavitation in superfluid ^4He

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We present the results of experiments to study cavitation in liquid ^4He at negative pressures. A focused sound wave is used to generate a negative pressure in a small region of the liquid, and the cavitation is detected by light scattering. We have studied the cavitation as a function of the temperature and static pressure applied to the liquid. We are able to observe the statistical nature of the cavitation process, and to determine the nucleation barrier and the attempt frequency. The results for the attempt frequency are lower than expected for homogeneous nucleation. We discuss the possibility that the nucleation is occurring on quantized vortices. The tensile strength at constant pressure decreases slowly with increasing temperature in the range 0.8 to 1.5 K, drops rapidly as the temperature approaches the λ point, and then decreases slowly above T_λ .

I. INTRODUCTION

Unlike a gas, a liquid is held together by cohesive forces, and thus, for short periods of time, like a solid, it can sustain positive stresses, i.e., negative pressures. However, after some length of time bubbles of the gas will form (cavitation), and the liquid will rupture, i.e., boil. The rate of bubble formation rises abruptly with negative pressure, and so there is a rather definite negative pressure at which cavitation becomes likely. The magnitude of this negative pressure is called the tensile strength of the liquid. Theories to describe the nucleation of bubbles by thermal fluctuations in an ideal pure liquid (homogeneous nucleation) have been worked out by several authors;¹⁻⁵ however, the tensile strength of most liquids is often much less than predicted by theory, because nucleation is seeded by walls, impurity particles, or dissolved gases (heterogeneous nucleation). Naturally, attention has turned to liquid ^4He , which can be made very pure, and in which nothing dissolves except ^3He . Nevertheless, early experiments in helium gave anomalously low values for the tensile strength.⁶⁻¹⁰ It seems likely that these early experiments were influenced by some form of heterogeneous nucleation. The experiments used large volumes of helium which probably contained particles of solid air, and positive and negative ions resulting from the passage of cosmic rays. In addition, in several of the experiments heterogeneous nucleation could have occurred on the walls of the helium container.

Recently, Nissen *et al.*^{11,12} performed an experiment in which ultrasound was generated by a hemispherical transducer. This produced a pressure oscillation at the acoustic focus which was thus confined to a small volume away from all surfaces. The use of a small helium volume greatly reduces the chance of heterogeneous nucleation, and it was found that the results agreed with the theory available at that time (see discussion below). Shortly after this, however, Maris and Xiong¹³⁻¹⁵ discovered

strong theoretical evidence that ^4He becomes unstable at -9 bars, and that the predictions of the earlier theories must be too high. They gave a corrected version of the theory, and then repeated Nissen's experiment using a similar apparatus.¹⁵ The new measurements gave a lower tensile strength than the experiments of Nissen *et al.*, and were lower than the modified theory by about a factor of 2.

In these experiments it is very difficult to determine an accurate value for the size of the pressure swing at the acoustic focus. It is likely that this is the cause of the discrepancy between the results of Nissen *et al.* and Xiong and Maris. In this paper we describe several experiments that we have performed that provide information about the cavitation process, but do not rely on an accurate pressure calibration. We have measured how the acoustic amplitude required to produce cavitation varies with temperature and with the static pressure applied to the liquid. In addition, we have studied the statistics of cavitation, and used the results of these studies to determine the height of the nucleation barrier and the value of the prefactor in the nucleation rate. The results for the prefactor suggest that quantized vortices may play a role in the cavitation process.

II. THEORETICAL BACKGROUND

A first-order phase transition is nucleated by the formation by thermal fluctuations of a microscopic seed, which then grows to macroscopic dimensions. However, the seed must exceed a certain critical size before this can happen. For the case of the liquid-gas transition, the standard approach is to assume that the free energy of a bubble can be written as

$$F = 4\pi R^2 \alpha - \frac{4\pi}{3} R^3 |P|, \quad (1)$$

where R is the radius of the bubble, α the surface energy,

and P the (negative) pressure in the liquid. While the second term represents a free-energy gain from converting liquid to gas (neglecting the saturated vapor pressure compared to $|P|$), the first term represents the energy cost of introducing the interface between the two phases. The result is that F has a maximum value $E = 16\pi\alpha^3/3|P|^2$ at the critical radius $R_c = 2\alpha/|P|$. Bubbles of radius less than R_c will be driven by thermodynamic forces to shrink rather than grow. Thus E represents an energy barrier to nucleation. The rate of nucleation Γ per unit volume and time thus has an activated form,

$$\Gamma = \Gamma_0 \exp(-E/k_B T), \quad (2)$$

where Γ_0 is a prefactor discussed below. If we apply a negative pressure throughout a volume V for a time τ , we may expect a cavitation event to become probable when $\Gamma V \tau > 1$, which leads to an estimate of the tensile strength⁵

$$P_t = \left[\frac{16\pi\alpha^3}{3k_B T \ln(\Gamma_0 V \tau)} \right]^{1/2}. \quad (3)$$

It can be seen from this result that the dependence of P_t on V and τ is remarkably weak, this being the basis for the remark in the introduction that one can consider the liquid to have a definite tensile strength.

For nucleation of bubbles in a classical liquid Fisher⁴ has given a discussion of the possible form of the prefactor Γ_0 . He considers the growth of the nucleus as a series of chemical reactions by which the size of the nucleus increases by one molecule. It is unlikely that this approach can be applied to nucleation in a superfluid. We return to a consideration of the prefactor in Sec. IV.

When this standard theory [Eq. (3)] is applied to liquid helium, the predicted tensile strength rises from around 7 bars at T_λ to ~ 15 bars at 0.4 K. Below this temperature it has been predicted⁵ that quantum tunneling through the nucleation barrier will dominate over thermal activation, and so the tensile strength will become independent of temperature. However, Maris and Xiong^{13,14} showed that the compressibility of liquid ⁴He diverges at a critical pressure $P_c \approx -9$ bar. The liquid is absolutely unstable below this pressure, and therefore it is impossible for the tensile strength to ever exceed $|P_c|$, i.e., the standard theory must fail at low temperatures where it predicts $P_t > |P_c|$. One can understand the source of the failure of the "standard theory" as follows. In writing down Eq. (1) it is assumed that the energy of the bubble can be divided into a surface term proportional to the surface energy, together with a term proportional to the volume and the applied pressure. This assumes that the radius of the bubble is large compared to the width of the interface. If the compressibility is large, however, the energy cost of making an interface with densities intermediate between the liquid and the gas is small, and the interface may be broad, indeed, comparable in width to the size of the critical radius. Maris and Xiong^{13,14} used a density functional theory to take this effect into account, and recalculated the value of the energy barrier E and the tensile strength P_t . According to their calculation the tensile strength is

$\sim 5-6$ bars at T_λ and increases slowly towards $|P_c|$ as $T \rightarrow 0$ K. In the Maris-Xiong theory the energy barrier is calculated using a temperature-independent density-functional scheme, and so the result for the barrier is independent of T . The effect of temperature on the nucleation barrier has been included in a recent calculation by Guilleumas *et al.*¹⁶ Their results give a tensile strength which is about 20% lower than the Maris-Xiong predictions at T_λ , but only 5% reduced at 1.5 K.

Even if the liquid is free of ions and solid impurities it may still contain quantized vortices. Because the circulation of liquid around a vortex line leads to a negative pressure, it is natural to consider the possibility that vortices act as sites for the heterogeneous nucleation of bubbles. Using a density-functional approach it is found that a straight vortex line becomes unstable against a uniform radial expansion at a critical pressure P_v .¹⁵ A simple density-functional scheme gives P_v in the range -6 to -7 bar, and a calculation by Dalfovo¹⁷ with a more sophisticated model gives -8 bar. Recently, Maris and Balibar^{18,19} have calculated the nucleation barrier for the formation of a bubble on a vortex. As expected, they find that at all pressures it is less than the nucleation barrier in vortex-free bulk liquid, and that it goes to zero smoothly as $P \rightarrow P_v$.

III. APPARATUS

The apparatus (Fig. 1) is similar to those used by Nissen *et al.*^{11,12} and Xiong and Maris.¹⁵ We pass ultrasound through the liquid thereby producing cavitation during the part of the oscillation when the pressure becomes negative. The transducer was a hemispherical piezoelectric-ceramic²⁰ (lead zirconate-titanate PZT) of inner radius 0.8 cm and wall thickness 0.2 cm immersed in the liquid and driven in a vibrational mode at 1.013 MHz. The concave surface of the transducer was directed downward, and a small hole was drilled vertically through the transducer at its center. The acoustic waves are focused into a small volume on the order of a half wavelength in size. The advantage of this geometry is that the small volume reduces the chances of heterogeneous nucleation, and the fact that the large pressure oscillations occur away from the transducer prevents the nucleation of bubbles at the surface. When the liquid is above the λ point the energy dissipation in the transducer

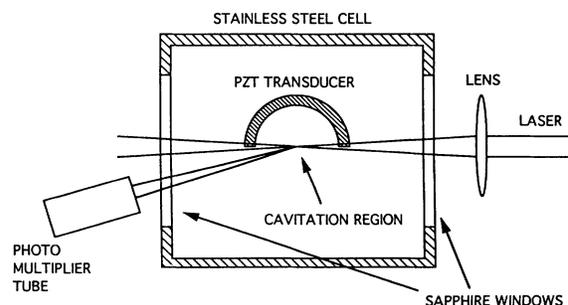


FIG. 1. Schematic diagram of the apparatus used to study cavitation.

can heat the helium immediately underneath the transducer surface. If a bubble is somehow formed in this region, the bubble will grow and eventually disrupt the operation of the transducer. The small hole drilled through the transducer allows a bubble to escape into the region above the transducer.

It is very difficult to make an accurate determination of the pressure swing at the acoustic focus in this experiment. As a first approximation, one could consider that the transducer was driven in its thickness mode. However, we suspected that there might be a strong coupling between thickness vibrations and flexural modes. To investigate this possibility we used a commercial software package²¹ to calculate the normal modes of the transducer. The results of this calculation showed that in the vicinity of 1 MHz the modes were complex mixtures of thickness and flexural vibrations. The elastic properties of PZT are dependent to a significant extent on the details of the manufacturing process, and the uncertainty in the elastic coefficients made it impossible to make a reliable calculation of the vibrational pattern associated with the particular normal mode chosen, or even to identify the mode. In the vicinity of 1 MHz the spacing of the normal modes was on the order of 20 kHz. The frequency 1.013 MHz was chosen simply because at this frequency the voltage drive to the transducer that was required to produce cavitation was a minimum.

As in the experiment of Xiong and Maris,¹⁵ the transducer was located in a cell that was filled via a capillary passing through the main helium reservoir. This filling procedure should eliminate frozen particles of air from the sample. We performed experiments both with ⁴He containing the natural abundance of ³He, and with isotopically purified ⁴He, but no difference in the results was detected. The volume of the cell was 300 cm³. The cell was attached to the pot of a pumped ⁴He cryostat with optical access and capable of reaching 0.82 K. The temperature of the pot was regulated by feedback from a carbon resistance thermometer, and the temperature of the cell was measured with a germanium resistance thermometer immersed in the liquid in the cell. A room-temperature mechanical gauge connected to the cell measured the static pressure, which could be varied up to the melting pressure.

To excite the transducer we used a low-level gated oscillator to produce a pulse of 700- μ s duration, which was amplified before being applied to the ceramic. Voltages as large as 60 V could be applied to the transducer, and this was sufficient to produce cavitation throughout the temperature range studied. Consequently, it was not necessary to use any impedance matching network between the rf amplifier and the transducer.

Cavitation was detected optically, but could also be seen by eye under some conditions. Light from a 10-mW helium-neon laser was focused by a 30-cm focal length lens onto the acoustic focus. The scattered light was detected with a photomultiplier tube (PMT) (response time $\sim 10 \mu$ s). We looked for an increase in the scattered light as a signature of cavitation, unlike Xiong and Maris¹⁵ and Nissen *et al.*^{11,12} who mainly used the change in transmitted intensity as a signal of cavitation.

A digital storage oscilloscope, from which the data could be read by a computer, registered both the excitation signal and the PMT response.

IV. RESULTS AND DISCUSSION

Because of the uncertainties in calculating the pressure swing at the acoustic focus we will present most of our discussion of the results in terms of the voltage applied to the transducer.

A. Qualitative observations

In the first experiments we studied superfluid helium at saturated vapor pressure. When a small voltage drive was applied to the ultrasonic transducer light scattering could be detected in the forward direction (deflection angle ≤ 5 mrad). We assume that this light has been scattered by the density fluctuations near to the acoustic focus. When the voltage was increased above a threshold level the intensity of light scattered in the forward direction increased and we were also able to detect scattered light at large angles (e.g., at 90° to the laser beam). The scattered light could easily be seen by eye. The threshold voltage at which the extra scattering appears varied with temperature. We assume that this light was scattered by cavitation events.

The scattered light appeared to come from a cloud surrounding the acoustic focus. The size and shape of this cloud varied randomly from pulse to pulse. The size of the cloud was of the order of 1 mm. No scattered light was detected until about 150 μ s after the beginning of the drive to the ultrasonic transducer. We assume that this delay comes from the transit time of the sound from the transducer to the focus together with the time for the oscillation of the transducer to build up to its full amplitude. Note that the onset of the scattering from the cloud occurred *before* second sound waves could arrive at the focus from the transducer; this provides evidence that the heat dissipated in the transducer is not causing the cavitation. The scattering from the cloud persisted for several hundred μ s after the acoustic drive was turned off.²² When the laser was focused onto the region below the acoustic focus the scattering from the cloud as detected by the PMT appeared at a time delayed with respect to the time of application of the voltage to the transducer. This indicated that the cloud drifted downwards (away from the transducer) with a velocity of about 1 m s⁻¹.

The scattering of the light from the cloud was observed to be much weaker at large angles (e.g., at 90°) than near to the forward direction (within ~ 50 mrad). The ratio of the intensity per solid angle was in the range 10^{-3} – 10^{-4} .

To understand these results one needs a theory of the growth and collapse of the cavitation bubbles. Some calculations of the dynamics of bubbles in normal liquid helium under ultrasonic excitation have been performed by Finch and Neppiras,²⁵ but unfortunately, as far as we are aware, a theory for bubbles in a superfluid has not been developed. The collapse of a bubble in a superfluid is likely to be significantly affected by the high speed with

which heat can be transported in the liquid, which means that the latent heat released when vapor condenses at the wall of a collapsing bubble is less likely to raise the wall temperature. In this context, it is interesting that the scattering persists for so long after the acoustic drive is turned off. If the rate at which the latent heat of the condensing vapor is transported away into the liquid is sufficiently large, and we can neglect the compressibility of both liquid and vapor, the collapse of the bubble is limited only by the inertia of the liquid. The time for a bubble of radius R to collapse as a result of external pressure P is²⁶

$$\tau = 0.915 \left[\frac{\rho R^2}{P} \right]^{1/2}, \quad (4)$$

where the liquid is taken to be incompressible with density ρ , and the effects of vapor inside the bubble are ignored. (The surface tension tends to make the bubble collapse even faster, but for bubbles at a depth of 3 cm below the surface, the hydrostatic head is greater than the Laplace pressure $2\alpha/R$ if $R > 20\mu$.) According to this formula, in order for a bubble to survive for 200 μ s at a depth of 3 cm, its starting radius must be at least 100 μ . This is a surprising result since, taken at face value, it means that the "cloud" that we see must actually be made up of a very small number of large bubbles, or possibly just one bubble. It is conceivable that even a *single* bubble could appear to the eye, and to a camera with a 1-ms exposure time, as a cloud if it were undergoing a sufficiently rapid random motion in the sound field. However, the angular dependence of the scattered light appears to rule out the idea that the cloud contains just a few large bubbles. A single large bubble (sufficiently large that geometrical optics can be applied) would reflect an amount of light from its surface proportional to $(n-1)^2/(n+1)^2$, where $n=1.028$ is the refractive index of liquid helium. In addition, light passing through the bubble will be refracted. In the geometrical optics approximation the maximum angle of light deflection is

$$2 \left[\frac{\pi}{2} - \sin^{-1} \left[\frac{1}{n} \right] \right] = 0.46 \text{ rad}. \quad (5)$$

However, most of the light should be refracted through angles smaller than the characteristic angle $2(n-1)=0.05$ rad. One finds that for a large bubble the ratio of the scattering I_1 at 90° to the scattering I_0 near the forward direction is²³

$$I_1/I_0 = (n-1)^4/2 = 3 \times 10^{-7}. \quad (6)$$

In the experiment I_1/I_0 is found to be in the range 10^{-3} – 10^{-4} , and is thus much larger than would be expected for scattering from a single large bubble.

To increase the relative intensity of the light scattered at large angles it appears necessary to propose that there are a large number of small bubbles (size comparable to or less than the light wavelength) that contribute to the scattering at large angles.²⁴ If this is the explanation it is necessary for there to be some process that enable small bubbles to survive for a few hundred μ s. One possibility

is that their rate of collapse is limited by the speed with which the vapor they contain can recondense at the walls. We have not attempted an analysis of this process. Another possibility is that, although the sound drive has been turned off, the helium cell still contains acoustic waves of an amplitude sufficient to affect the bubbles. The damping of a 1 MHz sound in helium in the temperature range 1–2 K during a time of 1 ms is negligible; the sound energy will become uniformly distributed throughout the cell and the amplitude will decrease slowly due to dissipation and sound transmission into the cell walls. The total acoustic energy introduced into the cell per pulse is of the order of 10^4 erg, and this means that after the sound is uniformly distributed the magnitude of the acoustic pressure fluctuations will be of the order of 10^{-2} bar. This is about 25 times larger than the hydrostatic head that drives the bubble to collapse. Thus, the acoustic field may continue to have a large effect on the dynamics of the bubbles.

Application of a static pressure to the helium had a dramatic effect on the light scattering:

(1) The light scattering at large angles decreased rapidly with increasing pressure, and at ~ 0.15 bar became too small to detect. The decrease was at least a factor of 10.

(2) The threshold voltage increased, as will be described below.

(3) The time dependence of the intensity of the scattered light with time was changed when pressure was applied. At saturated vapor pressure the scattered intensity near to the forward direction varied irregularly with time, and lasted somewhat longer than the duration of the acoustic pulse. At pressures above 0.4 bar, the cavitation signal appeared as well-defined spikes on top of an approximately constant background whose length was the same as that of the acoustic pulse. Figure 2 shows typical signals obtained under these conditions. The po-

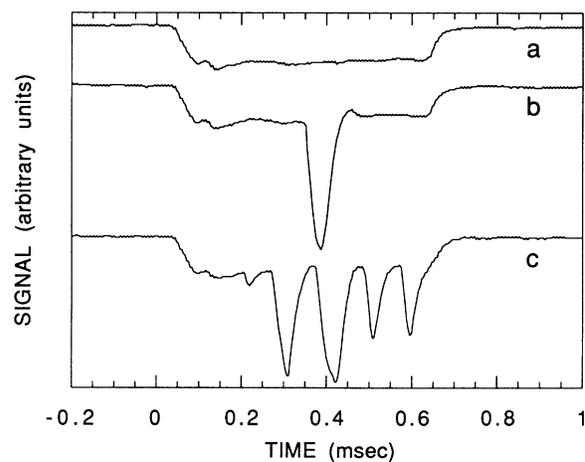


FIG. 2. Photomultiplier recording of the scattering of light at an angle of 0.011 rad as a function of time. The intensity increases going downwards on the plot. The data were taken at a static pressure of 1 bar. The three traces were recorded for the same drive voltage to the transducer. (a) shows scattering from the density fluctuation at the acoustic focus but no cavitation, whereas (b) and (c) show cavitation.

sition of the first spike relative to the beginning of the acoustic pulse was random. Sometimes there were multiple spikes, which were often approximately periodic in time [Fig. 2(c)]. As the ambient pressure is further increased, the spikes become smaller in amplitude and duration (perhaps because the bubble lifetime is approaching the response time of the PMT).

B. Observations of the statistics of the cavitation process

We noticed that for voltages in the vicinity of the threshold, cavitation occurred during some acoustic pulses but not others, even though the drive voltage was the same. We investigated the possibility that this was due to the fundamental statistical nature of the nucleation process. For a series of drive voltages and temperatures we measured the probability S of occurrence of cavitation, and the results are shown in Fig. 3. These data were taken at a static pressure of 1 bar. To determine the probability we looked at the light scattering from 100 acoustic pulses, and counted the number of times that a signal characteristic of cavitation was observed. In these measurements the amplitude of the pulse applied to the transducer was constant to within ± 50 mV, and the variation in amplitude from pulse to pulse was at a similar level. The stability of the temperature was typically ± 3 mK. There was no dependence of the probability of cavitation on the rate of repetition of the acoustic pulses which was varied from 1 per 1.5 s to 1 per 10 s.

To relate these results to theory we note that if one looks for cavitation in an experimental volume V_{exp} for a time τ_{exp} the probability of cavitation occurring is

$$S = 1 - \exp[-\Gamma_0 V_{\text{exp}} \tau_{\text{exp}} \exp(-E_{\text{exp}}/k_B T)], \quad (7)$$

where E_{exp} is the value of the energy barrier corresponding to the pressure in the experimental volume. Our ex-

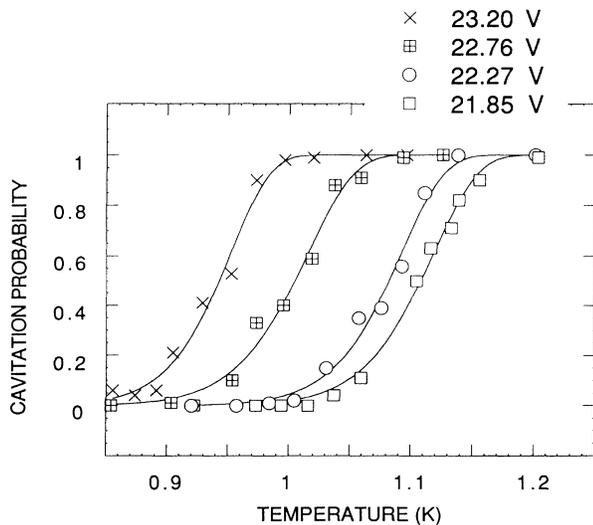


FIG. 3. Probability S of cavitation as a function of temperature for four different drive voltages.

periment is complicated by the fact that the pressure field varies in space and time. There is a maximum negative pressure P_{min} that is reached at the acoustic focus once during each cycle of the sound wave. At this point in space and time the nucleation barrier takes on its minimum value E_{min} . The experimental volume is roughly the volume of the liquid in which the barrier is no more than $k_B T$ greater than E_{min} , and the experimental time can be thought of in a similar way. A more quantitative analysis can be given as follows. Let us suppose that for a particular cycle of the pressure oscillation the minimum pressure occurs at $t=0$. We suppose that near to the space-time point of minimum pressure we can approximate the pressure as

$$P(r, t) \approx P_{\text{min}}(1 - ar^2)(1 - bt^2), \quad (8)$$

where a and b are constant coefficients, and r is the distance from the point where the pressure is minimum. Then the probability that cavitation will occur at some point during the sound pulse is

$$S = 1 - \exp\left[-N \int dV \int dt \Gamma_0 \exp[-E(P)/k_B T]\right], \quad (9)$$

where N is the number of cycles in the acoustic pulse, and $E(P)$ is the barrier height when the pressure is P . We make the approximation that E varies linearly with P , and that the variation of Γ_0 with pressure can be neglected in this context. Then one can perform the integrals to obtain

$$S = 1 - \exp\left[-\frac{\pi^2 N \Gamma_0 k_B^2 T^2}{(d \ln E / d \ln |P|)^2 E^2 a^{3/2} b^{1/2}} \times \exp(-E_{\text{min}}/k_B T)\right]. \quad (10)$$

If we approximate the spatial variation of P by the function $\sin(kr)/kr$, where k is the acoustic wave number, we obtain

$$a = \frac{2\pi^2}{3\lambda^2}, \quad (11)$$

where λ is the sound wavelength. Similarly, taking the time variation of the pressure as proportional to $\cos(2\pi t/\tau)$, where τ is the sound period, gives $b = 2\pi^2/\tau^2$. If these values for a and b are inserted into Eq. (10) and the result compared to Eq. (5), we obtain

$$V_{\text{exp}} \tau_{\text{exp}} = \frac{3^{3/2} N \lambda^3 \tau k_B^2 T^2}{4\pi^2 (d \ln E / d \ln |P|)^2 E^2}. \quad (12)$$

In this expression we have replaced E_{min} by E to simplify the notation. To make a fit to the data we note that our result for $V_{\text{exp}} \tau_{\text{exp}}$ is proportional to T^2 . In addition, based on the Fisher theory⁴ the prefactor Γ_0 is proportional to T . (We discuss this point below.) Consequently, we have fit our results for S to the function

$$S = 1 - \exp[-AT^3 \exp(-E/k_B T)], \quad (13)$$

where A and E are adjustable parameters, and the prefactor Γ_0 is related to A via

$$\Gamma_0 = \frac{AT(d \ln E / d \ln |P|)^2 E^2 a^{3/2} b^{1/2}}{\pi^2 N k_B^2} \quad (14)$$

The results for A and E are listed in Table I, together with the drive voltage V and the temperature $T_{1/2}$ at which the measured probability S had a value of $\frac{1}{2}$. To determine Γ_0 from A we have to use a value for $d \ln E / d \ln |P|$. If we assume that the nucleation of cavitation is occurring in the bulk of the liquid, then we can take the value of this derivative from the theory of Xiong and Maris.^{13,14} For energies in the range around 30 K, this theory predicts that $d \ln E / d \ln |P|$ is approximately 7. Using this value we obtain the results for Γ_0 that are included in Table I. Given these results for Γ_0 and E we checked the validity of the Gaussian approximation that we used to evaluate the integral in Eq. (9), and found that it was well-justified. We estimate that the uncertainty in $\log_{10} \Gamma_0$ is about 1.5. We remark that the uncertainties quoted for E and Γ_0 are highly correlated; the combination $E / (k_B \ln \Gamma_0 V_{\text{exp}} \tau_{\text{exp}})$ is a temperature which in effect determines the position of the transition in the data in Fig. 3, and the estimated statistical uncertainty in this quantity is only 0.003 K.

The results show that the prefactor lies in the range $10^{25} - 10^{27} \text{ cm}^{-3} \text{ s}^{-1}$. Rather than attempting a comparison with the theory of the prefactor due to Fisher, which does not appear to be relevant to a superfluid, it is more constructive to consider the problem from first principles. The liquid contains phonons which will cause the local pressure to fluctuate. At temperature T it is straightforward to show that the rms pressure fluctuation has a magnitude

$$\langle (\delta P)^2 \rangle^{1/2} = 0.79 T^2 \text{ bar}, \quad (15)$$

where T is measured in K. This formula is based on the assumption that the pressure swings are sufficiently small that δP can be considered to be proportional to $\delta \rho$. If there is a fluctuation such that the pressure exceeds the instability pressure P_c over a volume of the size of the critical nucleus, then nucleation will occur. Pressure fluctuations that extend over smaller volumes will not be able to produce cavitation, even if the pressure exceeds P_c . The prefactor is thus the rate of occurrence of statistically independent fluctuations that create the required pressure swing over the required volume. For pressure swings produced by thermal phonons, the pressure at a

particular point in space is correlated with the pressure at the same point a time τ later if $\langle \nu \rangle \tau$ is less than 1, where $\langle \nu \rangle$ is the average frequency of a thermal phonon. Thus, we can consider that in the time-domain a "new" pressure fluctuation is produced at a rate of $\langle \nu \rangle$ per unit time. The pressure fluctuations are correlated in space over a distance scale set by the wavelength of a thermal phonon; for a typical phonon of energy $3k_B T$ this wavelength is²⁷

$$\langle \lambda \rangle = \frac{hc}{3k_B T} = \frac{38}{T} \text{ \AA}, \quad (16)$$

where c is the sound velocity. Since λ is similar in size to the dimensions of the critical nucleus, the pressure fluctuations typically produced by the phonons are over a length scale that is sufficiently large to produce cavitation. The pressure fluctuations are uncorrelated over distances greater than $\langle \lambda \rangle$; thus the number of independent fluctuations per unit volume is $\langle \lambda \rangle^{-3}$. In this way we obtain an estimate for the prefactor of

$$\Gamma_0 = \langle \nu \rangle / \langle \lambda \rangle^3 = (3k_B T / h)^4 / c^3. \quad (17)$$

If one were to consider a situation in which the size of the critical nucleus was larger than $\langle \lambda \rangle$, then clearly only the fluctuations due to the low-frequency part of the phonon spectrum should be considered, and this would modify Eq. (17). At 1 K the value of Γ_0 given by this formula is $2 \times 10^{30} \text{ cm}^{-3} \text{ s}^{-1}$, which is about 5 orders of magnitude larger than our experimental results, and significantly outside the range of our experimental uncertainty. This suggests that we may be detecting heterogeneous, rather than homogeneous nucleation in the bulk of the liquid. If we suppose that this nucleation occurs on vortices the prefactor would become

$$\Gamma_0 = \Gamma_{0,\text{vort}} L_{\text{vort}}, \quad (18)$$

where L_{vort} is the length of vortex line per unit volume, and $\Gamma_{0,\text{vort}}$ is the attempt rate per unit length of vortex line and per unit time. If we apply the same ideas as above to the consideration of the prefactor we would have

$$\Gamma_{0,\text{vort}} = \langle \nu \rangle / \langle \lambda \rangle = (3k_B T / h)^2 / c. \quad (19)$$

Thus at around 1 K Γ_0 is around $1.5 \times 10^{17} L_{\text{vort}} \text{ cm}^{-3} \text{ s}^{-1}$, with L_{vort} in cm^{-2} . Thus, to explain the experimental results for the prefactor one would have to suppose that the vortex line density was in the range $10^8 - 10^{10} \text{ cm}^{-2}$. This high density of vorticity might be produced by the ultrasound itself. Schwarz and Smith²⁸ have shown that vortex densities of 10^4 cm^{-2} are generated by ultrasound at intensities of only a few mW/cm^2 . The pressure swings in our experiment are in the range of several bars, and a rough estimate indicates that the ultrasound in our experiment reaches intensities of kW/cm^2 in the focal region.

It is important to note a limitation of this analysis that we have given. We have implicitly assumed that the height of the energy barrier can be considered to be independent of temperature. Without this assumption it would be impossible to determine both the barrier height

TABLE I. The nucleation barrier E and the prefactor Γ_0 as determined from the variation with temperature of the probability S of cavitation. V is the voltage applied to the transducer, $T_{1/2}$ the temperature at which the probability $S = \frac{1}{2}$, and A is the fitting parameter in Eq. (13).

V (volt)	$T_{1/2}$ (K)	A (K ⁻³)	E/k_B (K)	Γ_0 (cm ⁻³ s ⁻¹)
23.2	0.95	1.4×10^{12}	26.6 ± 3	$10^{25.6 \pm 1.5}$
22.8	1.02	1.1×10^{11}	25.9 ± 3	$10^{24.5 \pm 1.5}$
22.3	1.10	7.9×10^{12}	32.8 ± 4	$10^{26.6 \pm 1.5}$
21.9	1.13	5.9×10^{12}	33.4 ± 4	$10^{26.5 \pm 1.5}$

and the prefactor. The assumption of a temperature-independent barrier appears reasonable in the temperature range where we have carried out the analysis of $S(T)$, i.e., below 1.13 K. The temperature dependence of the surface energy and the sound velocity is very small in this temperature range. However, it is clear that we cannot use the analysis of $S(T)$ to study the barrier at temperatures close to T_λ , or in the normal fluid.

C. Temperature dependence of the cavitation threshold

If the cavitation is indeed occurring on quantized vortices, it is of interest to study the change in the cavitation strength in the vicinity of the λ point. Quantized vortices have only been observed in superfluids, and so we expected that there might be a sharp increase in the tensile strength on raising the temperature through T_λ .²⁹ To test this idea we made measurements of the temperature dependence of the cavitation threshold V_c , defined as the voltage required to give a cavitation probability of $\frac{1}{2}$. Figure 4 shows the dependence of V_c on temperature at fixed density for four different densities. There is a marked drop as the λ point is approached from below. Above T_λ the drive voltage increases with increasing temperature. We have indicated in Fig. 4 the λ transition temperature for the density corresponding to each curve. A change in slope of V_c vs T is seen to lie close to the λ point at each density. This close correspondence is perhaps rather surprising. The cavitation presumably occurs at the peak of the negative pressure swing associated with the sound wave. This pressure swing will displace the liquid in the P - T plane by a significant distance, primarily parallel to the P axis but there will also be a change in the temperature. Thus, if the static pressure and temperature lie on the λ line, it is not clear that the liquid will still be close to the λ line when the cavitation actually occurs. The fact that there is a change in slope of V_c vs T when the static pressure and temperature are on near the λ line is therefore an interesting result that needs further investigation.

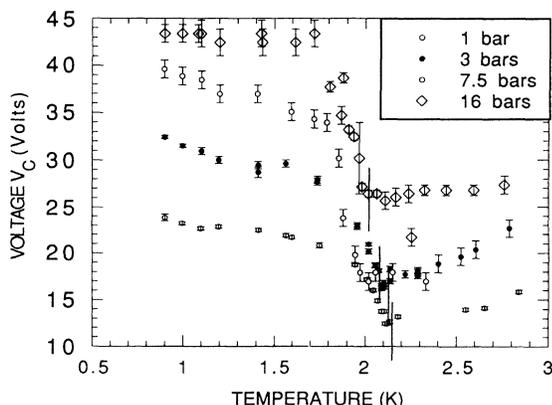


FIG. 4. Voltage threshold V_c for producing cavitation as a function of temperature at fixed density. The pressures indicated in the figure are the pressure in the cell at 0.9 K. The vertical bars indicate the location of the λ point for each density.

We have also measured the dependence of V_c on the static pressure P_{st} . Data taken at 1.2 K are shown in Fig. 5. V_c initially rises linearly with the ambient pressure, and then levels off. The results obtained at two other temperatures (0.9 and 2.3 K) were qualitatively similar. The signals became progressively harder to see at higher ambient pressures. The character of the waveform of the scattered light signal changed continuously as the pressure was raised. This suggests that even at high static pressures we are still observing the cavitation of bubbles, rather than the nucleation of solid during the positive part of the pressure swing.

The initial slope dV_c/dP_{st} of the curve of cavitation voltage vs static pressure was observed to be almost independent of temperature. We used this derivative to adjust the results for V_c vs T along the lowest density isopycnal to deduce the dependence of V_c on temperature at a constant pressure of 1 bar. The result is shown in Fig. 6. This result for V_c still decreases rapidly as one approaches the λ point from below. Above T_λ , V_c continues to decrease, in contrast to the increase that occurs along the isopycnals. The drop in tensile strength at the λ point has also been observed by Nissen *et al.*¹² We have considered several possible explanations for the rapid variation of the cavitation strength in the range just below T_λ . The surface energy α of helium has a small anomaly at T_λ and this must affect the nucleation rate and cavitation strength. However, the anomaly is so small³⁰ that it is unlikely that the effect that we see can be explained in this way. The sound velocity also has an anomaly at T_λ , and this presumably leads to a temperature dependence of the pressure P_c at the limit of stability. The temperature dependence of P_c and the effect of this on the nucleation rate has not been calculated. The acoustic impedance of helium varies with temperature, and so the coupling of the transducer to the liquid is not constant; but it appears that this would raise the voltage threshold near T_λ rather than lower it. It may be significant that the drop in tensile strength begins well below the λ point, and that there appears to be no discontinuity at T_λ . This result indicates that the effect is due

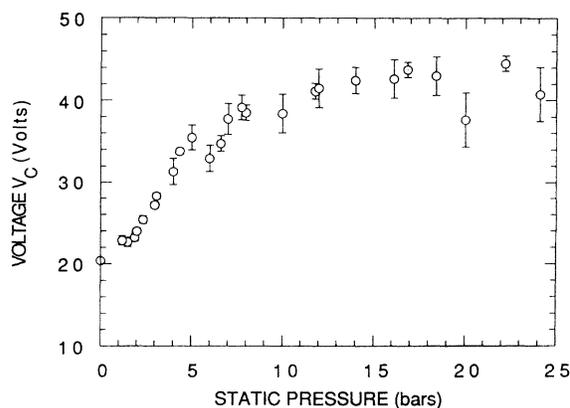


FIG. 5. Threshold voltage required to produce cavitation as a function of the static pressure applied to the liquid. These data were taken at 1.2 K.

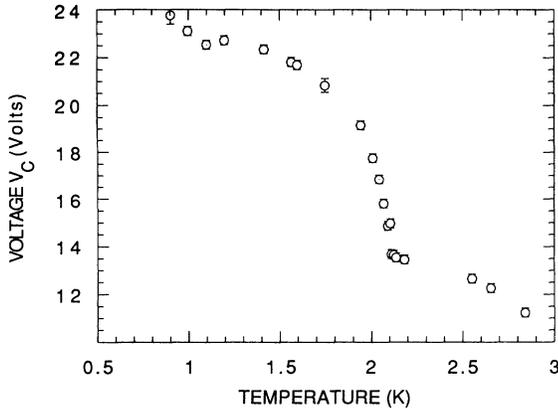


FIG. 6. Voltage threshold V_c for producing cavitation as a function of temperatures at a fixed pressure of 1 bar. The threshold has been calculated by applying a correction to the isopycnal data shown in Fig. 4.

to some property of the superfluid which approaches the normal value as the λ point is approached from below. If we accept the evidence from the prefactor determination that we are seeing heterogeneous nucleation on vortices, it would be necessary either for the line density of vortices to increase rapidly as T_λ is approached, or for the energy barrier for nucleation on a vortex to undergo a large decrease. A rapid increase in L_{vort} as $T \rightarrow T_\lambda$ has been predicted by Williams.³¹ He also predicts that quantized vortices are present in normal liquid helium, and thus a development of his theory might be consistent with our result that there is no discontinuity in the cavitation strength at T_λ . However, it is important to note that Williams' theory considers the thermal equilibrium density of vortex line, while in our experiments it seems

likely that the presence of the sound wave greatly increases the vortex density. One could perhaps determine the density of vortices in the focal region by measuring their interaction with a second sound pulse.

V. SUMMARY

In this paper, we have demonstrated the thermally activated nature of the cavitation process in liquid helium near 1 K, and we have been able to observe the statistical nature of the cavitation process. By measuring the cavitation probability as a function of temperature we have been able to determine both the activation energy E and the prefactor Γ_0 . This is in contrast to measurements of the tensile strength alone, which can only give a combination of the activation energy and the prefactor. The results for Γ_0 are significantly smaller than the values expected for homogeneous nucleation, and this suggests that this heterogeneous nucleation may be occurring on quantized vortices. However, we find that the cavitation strength has no discontinuity at the λ point, and hence for vortices to be the explanation of our results it is necessary to suppose that vortices continue to exist in the normal phase.

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¹M. Volmer and A. Weber, *Z. Phys. Chem.* **119**, 277 (1926).

²R. Becker and W. Doring, *Ann. Phys.* **24**, 719 (1935).

³Ya. B. Zel'dovich, *Zh. Eksp. Teor. Fiz.* **12**, 525 (1942).

⁴J. C. Fisher, *J. Appl. Phys.* **19**, 1062 (1948).

⁵V. A. Akulichev and V. A. Bulanov, *Akust. Zh.* **20**, 640 (1974) [*Sov. Phys. Acoust.* **20**, 501 (1975)].

⁶J. W. Beams, *Phys. Rev.* **104**, 880 (1956).

⁷R. D. Finch, R. Kawigada, M. Barmatz, and I. Rudnick, *Phys. Rev.* **134**, A1425 (1964).

⁸P. D. Jarman and K. J. Taylor, *J. Low Temp. Phys.* **2**, 389 (1970).

⁹P. L. Marston, *J. Low Temp. Phys.* **25**, 383 (1975).

¹⁰H. J. Maris, S. Balibar, and M. S. Pettersen, in *Proceedings of the Conference on Vorticity and Bose-Condensates, Minnesota, 1993* [*J. Low Temp. Phys.* **93**, 1069 (1993)]. This paper lists most of the early references to cavitation in liquid helium, and includes a preliminary account of the experimental results discussed in the present paper.

¹¹J. A. Nissen, E. Bodegom, L. C. Brodie, and J. S. Semura, *Adv. Cryogen. Eng.* **33**, 999 (1988).

¹²J. A. Nissen, E. Bodegom, L. C. Brodie, and J. S. Semura,

Phys. Rev. B **40**, 617 (1989).

¹³H. J. Maris and Q. Xiong, *Phys. Rev. Lett.* **63**, 1078 (1989).

¹⁴Q. Xiong and H. J. Maris, *J. Low Temp. Phys.* **77**, 347 (1989).

¹⁵Q. Xiong and H. J. Maris, *J. Low Temp. Phys.* **82**, 105 (1991).

¹⁶M. Guilleumas, M. Pi, M. Barranco, J. Navarro, and M. A. Solis, *Phys. Rev. B* **47**, 9116 (1993).

¹⁷F. Dalfovo, *Phys. Rev. B* **46**, 5482 (1992).

¹⁸H. J. Maris and S. Balibar, *Proceedings of the LT20 Conference, Eugene, Oregon, 1993* [*Physica B* (to be published)].

¹⁹H. J. Maris, *J. Low Temp. Phys.* **94**, 125 (1994).

²⁰Manufactured by Quartz et Silice, France, type P7-62.

²¹We used the ABACUS mechanical engineering software package developed by Hibbit, Karlsson, & Sorensen, Inc., Providence, Rhode Island.

²²This time was significantly longer than the ring-down time of the ultrasonic transducer.

²³The derivation of this formula assumes scattering of unpolarized light and treats $(n - 1)$ as a small parameter.

²⁴The angular dependence of the scattering from a small particle varies as $(1 + \cos^2\theta)$.

²⁵R. D. Finch and E. A. Neppiras, *J. Acoust. Soc. Am.* **53**, 1402 (1973).

²⁶Lord Rayleigh, *Philos. Mag.* **34**, 94 (1917).

²⁷E. M. Lifshitz and L. P. Pitaevskii, *Statistical Physics, Part 2* (Pergamon, Oxford, 1980), Chap. 9.

²⁸K. W. Schwarz and C. W. Smith, *Phys. Lett.* **82A**, 251 (1981).

²⁹This has also been suggested by M. Guilleumas *et al.* in Ref. 16.

³⁰M. Iino, M. Suzuki, and A. Ikushima, *J. Low Temp. Phys.* **61**, 155 (1985).

³¹G. A. Williams, *Phys. Rev. Lett.* **59**, 1926 (1987); **68**, 2054 (1992).