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Nucleation of Bubbles in Liquid Helium

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We give a brief survey of experiments that have been performed to study the nucleation of bubbles in liquid helium at negative pressures.

1. INTRODUCTION

In this paper we give a brief summary of studies that have been made of the nucleation of bubbles (cavitation) in liquid helium under negative pressure. There have been two principal motivations for research in this field. Because all impurities (except ^{3}He) freeze out of the liquid at low temperatures, it is possible to prepare helium with a much higher purity than ordinary classical liquids. In any study of a nucleation process this is an important advantage because impurities introduce the complication of heterogeneous nucleation. The second reason for interest in helium is that at low enough temperatures nucleation is expected to be dominated by quantum tunnelling rather than thermal activation.

We begin with a discussion of the theoretical background. The early experiments, which were typically performed on relatively large volumes of helium are in striking disagreement with homogeneous nucleation theory, and several possible explanations for this have been proposed. In the last few years three sets of experiments have been performed in which the experimental conditions are better controlled than in the earlier measurements. The results of these experiments are in much closer agreement with theory, but there are still major unresolved questions.

2. THEORETICAL BACKGROUND

The standard approach to the nucleation of a bubble considers the energy required to form a bubble of radius R in bulk liquid. This energy is

$$E = 4\pi R^2\alpha + \frac{4}{3}\pi R^3 P \quad (1)$$

where α is the liquid-gas surface energy, and P is the applied pressure, which is negative. For small R this energy is dominated by the surface term and is positive, but for sufficiently large R, E becomes negative. The maximum value E_{max} of the energy is at $R = 2\alpha/|P|$, and is given by

$$E_{max} = \frac{16\pi\alpha^3}{3P^2} \quad (2)$$

This acts as a barrier against nucleation, and so if we assume that nucleation occurs by thermal activation, the nucleation rate $\Gamma(P,T)$ is given by

$$\Gamma(P,T) = \Gamma_0 \exp(-E_{max}/kT). \quad (3)$$

The prefactor Γ_0 can be considered to be the number of independent nucleation sites per unit volume multiplied by an attempt frequency. It is important to note that $\Gamma(P,T)$ is a rate per unit volume and per unit time. Thus the probability S of nucleation occurring in a volume V after a waiting time τ is

$$S = 1 - \exp(-V\tau\Gamma(P,T)) = 1 - \exp[-V\tau\Gamma_0 \exp(-E_{max}/kT)] \quad (4)$$

As the pressure becomes more negative the height of the energy barrier decreases and the nucleation rate increases rapidly. The tensile strength P_n of the liquid is defined as the magnitude of the negative pressure at which S becomes of the order of 1. From the preceding three equations it follows that

$$P_n = \sqrt{\frac{16\pi\alpha^3}{3kT\ln(\Gamma_0 V\tau)}} \quad (5)$$

The surface tension of helium has been measured by Iino *et al* ,[2] who find the value 0.355 erg cm^{-2} as $T \to 0$ K. Using this value, and taking for the purposes of illustration $V\tau = 10^{-10}$ cm^3 sec and $\Gamma_0 = 10^{30}$ cm^{-3} sec^{-1}, Eq. 5 gives the tensile strength shown in Fig. 1. Note that because of the form of Eq. 5 there is only a weak dependence of the tensile strength on V and τ.

This argument predicts that at low temperatures the tensile strength should vary as $T^{-1/2}$. This increase in P_n as the temperature decreases is limited by two distinct physical processes. Even at $T = 0$ K there remains a finite nucleation rate due to quantum tunnelling through the energy barrier.[3] Within the model just described tunneling is predicted to become an important process in the temperature range below about 0.5 K where the tensile strength has risen to 15 to 20 bars.[4] The second correction that has to be included involves the equation of state of helium at negative pressures.[5] It has been shown[5] that the bulk liquid becomes macroscopically unstable at a critical pressure P_c of around − 9 bars, i.e. the sound velocity becomes zero and the liquid is unstable against long wavelength density fluctuations. Thus, a tensile strength larger than $|P_c|$ is impossible. A revised calculation of the tensile strength has been performed that allows for this effect, and the results are included in Fig. 1.

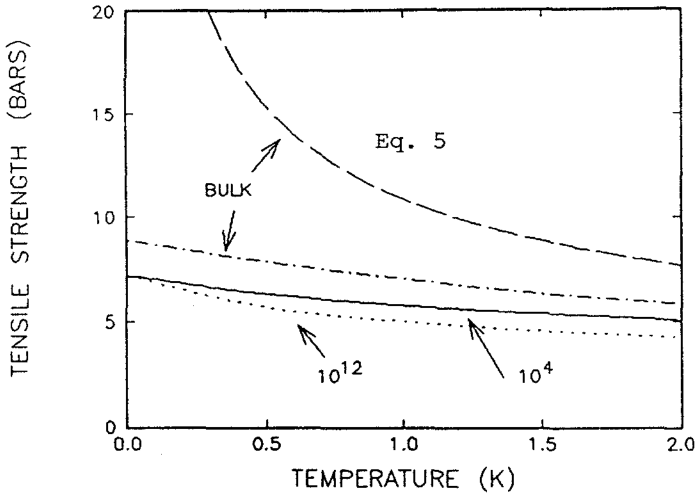

Fig. 1. Calculated tensile strength of liquid helium. The dashed curve shows the predictions of Eq. 5. The tensile strength when allowance is made for the instability of the bulk liquid at P_c is shown by the dashed-dotted line. The solid and dotted curves show the predicted tensile strength for liquid containing 10^4 and 10^{12} vortices per cm^2, respectively.

The results shown in Fig. 1 assume an energy barrier that is independent of temperature, i.e. the low temperature limiting value of the surface tension and the equation of state have been used. Recently, Guilleumas *et al*[6] have extended the calculations to allow for a temperature-dependent barrier. This results in a reduction of the tensile strength that is very small below 1 K, but which increases to about 20 % at 2 K.

3. EARLY EXPERIMENTS

Here we give a highly condensed summary of some of the early experiments on cavitation in liquid helium.

The first quantitative measurement of the tensile strength of helium was performed by Beams in 1956.[7] Helium was contained in a capillary that was spun about an axis perpendicular to its length, thereby setting the liquid into a state of tension. At a critical rotation speed the helium column in the capillary broke. The tensile strength at 1.8-1.9 K was found to be 0.14 bars. Beams noted that this value was more than one order of magnitude less than the theoretically expected result.

In the 1960's a large number of studies were carried out under a variety of conditions. Finch, Kagiwada, Barmatz and Rudnick[8] used a planar ultrasonic transducer operating at 90 kHz to apply an oscillating pressure to a large volume of helium (several cc). The magnitude of the oscillating pressure required to produce cavitation below the lambda point was found to be between 1000 to 2000 dynes cm^{-2}, and was in the same range as the *positive* hydrostatic pressure head due to the helium bath. Thus, the measured tensile strength was extremely small, i.e. of the order of 10^{-3} bar. Cavitation was detected via the acoustic emission associated with the collapse of cavitation bubbles, and also visually at the higher drive amplitudes. Below T_λ the tensile strength dropped with increasing temperature, but on raising the temperature above T_λ there was an abrupt increase in P_n. Finch and Wang[9] made measurements with a hollow cylindrical transducer and studied the thresholds for both audible and visible cavitation. Below T_λ the visible threshold was an order of magnitude higher than the audible threshold, but above T_λ the difference in the two thresholds was much less. Further measurements[10] studied the effect of the applied static pressure on the cavitation; this was found to raise the threshold for visible cavitation but not influence the audible threshold. Experiments by Jarman and Taylor[11] gave results that were in qualitative agreement with the earlier measurements by Finch. Thus, it was again found that the threshold for audible cavitation was in the range $10^{-3} - 10^{-4}$ bar, and a difference in the threshold for audible and visible cavitation was found below T_λ, and not above. Movies of the dynamics of cavitation bubbles have been made by Edwards *et al* ,[12] Mossé *et al*[13] and Marston.[14]

Interesting experiments to investigate the effect of stirring the helium were also performed by Finch *et al.*[15,16] A rod was inserted into the sample cell and rotated at frequency ω while the threshold for audible cavitation was measured. It was found that at a critical rotation frequency of the rod ω_c the cavitation threshold underwent a sudden drop. It was proposed that this drop was associated with the introduction of vortices into the liquid, and this idea was supported by further experiments by Dhingra and Finch.[17]

A wide variety of ideas have been put forward to explain these results, but there is still no definite understanding of what is happening in these experiments. It is important to note that the volumes of helium studied in these early experiments were in most cases large. Thus, experiments that use a planar ultrasonic transducer to generate a fluctuating pressure typically study a volume of several cc. Cylindrical transducers focus the sound to a region surrounding the cylinder axis, and so for a transducer of length $L \sim 1$ cm long operating at a frequency of 40 kHz, for example, the negative pressure swing occurs over a volume of the order of $L(\lambda/2)^2 \approx 0.1$ cc. Given that these volumes are large, it is certainly reasonable to suppose that heterogeneous nucleation could occur and that this may explain why the cavitation strength is so much lower than theory predicts. The helium could contain particles of frozen air (no special care was taken to keep the helium clean in most of the experiments), positive or negative ions generated by cosmic rays, and vortices (possibly produced by the sound field itself). However, even if one supposes the helium contains *all* of these defects it is not clear how this gives cavitation thresholds as low as those measured. Some possibilities are the following:
1) Solid Particles. If the helium contains a solid particle that the liquid does not wet, (i.e. the liquid has a non-zero contact angle θ), the barrier against nucleation via formation of a bubble that intersects the solid surface is less than the barrier for nucleation in the bulk.[1] However, to give a reduction of the tensile strength by the required factor one needs the contact angle to be close to 180° which appears unlikely.[18] The shape of the particle will also play a role in determining the reduction in the barrier height.

2) Ions. The effect of electron bubbles in the liquid has been estimated by Akulichev and Boguslavskii.[19] They find a reduction in the cavitation strength to a value of around 1 bar at 2 K, still much larger than the experimentally-measured values.
3) Cosmic Rays. Another possibility is that the cavitation is a result of the *energy* deposited by a cosmic ray, rather than by the ions left in the liquid. Ionizing particles have been used to induced cavitation in a large number of classical liquids,[20] and electrons in these liquids do not form bubbles. In the Glaser bubble chamber[21] the bubbles are believed to be formed via the deposition of energy by the secondary electrons produced along the track.[22] A theory of this mechanism of bubble formation for a superfluid has apparently not been developed, so it is not clear if this approach can explain the *increase* in the cavitation threshold observed on going from below T_λ to above. However, the viscosity of the liquid does have a significant effect on the dynamics of bubble formation and this provides one possible way in which the tensile strength could change drastically at the lambda temperature.
4) Vortices. The idea that the nucleation of bubbles might be associated with vorticity was proposed as long ago as 1944 by Dean,[23] and this proposal is supported by the experiments of Finch and coworkers.[15,16] It is clear that the circulation around a vortex will lower the energy that is required to form a bubble. However, it appears that this velocity field is *much* too weak to affect the nucleation barrier at low pressures. To see this recall that the radius R of the critical nucleus without a vortex is $2\alpha/P$. For a pressure of 10^{-3} bar this radius is thus 70,000 Å. The modification of the critical nucleus when a vortex is present (for example when a straight vortex passes through the center of the bubble) is very small because most of the surface of the critical nucleus is a long way from the vortex core and consequently the flow field is small. This result is confirmed by the calculation described in the next section.

4. EXPERIMENTS ON SMALL VOLUMES

Recently experiments have been performed in which very much smaller volumes of liquid are studied.[24,25] In these experiments a hemispherical transducer is used, whereas the earlier experiments used a planar or cylindrical transducer. The volume of liquid studied is now of the order of $(\lambda/2)^3$, where λ is the acoustic wavelength. In addition, the new experiments use a higher frequency (500 kHz to 1MHz), thus giving a shorter sound wavelength and a further substantial reduction in volume. This greatly reduces the probability that cosmic rays or other radioactive sources will interfere with the experiment. The first experiments were performed by Nissen *et al*[24] and covered the range of temperatures from 1.7 to 4 K. They found a tensile strength that was around − 8 bars at 1.7 K and decreased monotonically with increasing temperature. There was some evidence for a rapid drop in tensile strength just below T_λ. Xiong and Maris[25] performed a similar experiment in the temperature range 0.8 to 2 K. In this temperature range they found a tensile strength that decreased slowly and smoothly with increasing temperature. In the narrow temperature range where the two data sets overlapped (1.7 to 2 K), the tensile strengths measured in the two sets of experiments differed by about a factor of 3. This difference probably arises from the difficulty in determining the magnitude of the pressure swing in this type of experiment. Even if the piezoelectric and me-

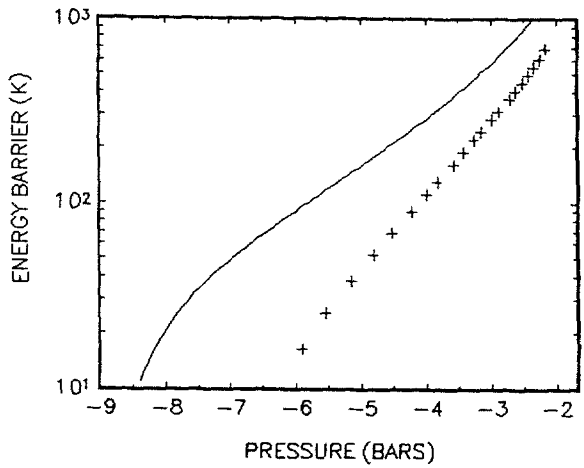

Fig. 2. Nucleation barrier for the formation of a bubble in liquid helium at low temperatures. The solid line shows the barrier in bulk liquid and the crosses are the results of calculations of the barrier for the formation of a bubble on a vortex.

chanical properties of the transducer can be reliably measured, it is still necessary to perform a complicated calculation in non-linear acoustics to follow the acoustic wave into the focus.

The fact that these experiments give a large tensile strength (in the range of bars) raises the possibility that homogeneous nucleation is indeed being observed. However, the potential influence of vortices is still of concern because the high amplitude sound waves that are used to generate the negative pressures can generate a large amount of vorticity.[26] Theoretical investigations[25,27,28] of the properties of a vortex at negative pressures show that at a critical negative pressure P_v a long straight vortex becomes unstable against a uniform radial expansion. This instability is reached before the instability of the bulk liquid at P_c. Estimates of P_v lie between − 6.5 and − 8 bars, depending on the microscopic model that is used.[25,27,28] In addition, a calculation has been performed of the nucleation barrier for the formation of bubbles on a vortex line.[28] The energy barrier is, as expected, less than the barrier in bulk helium. The results for the energy barrier are shown in Fig. 2, along with the energy barrier in bulk liquid calculated by Maris and Xiong.[5] The predicted change in the tensile strength due to the presence of vortices is included in Fig. 1.

Very recently we have made more precise measurements of the temperature-dependence of the cavitation threshold and have been able to observe the statistical nature of the nucleation process.[29,30] The experiments use a hemispherical transducer, together with improved optics for detection of light scattered from bubbles. To observe the statistics of cavitation a series of electrical driving pulses each of the same size was applied to the transducer while the temperature of the helium was

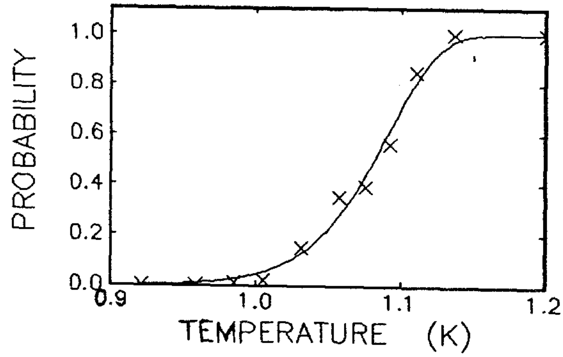

Fig. 3. Measurements of the probability of nucleation of a bubble as a function of temperature. The drive to the transducer is held constant during this series of measurements. The solid curve is the result of a fit using Eq. 4 with the prefactor and the height of the nucleation barrier taken as fitting parameters.

held constant. For each drive pulse it was determined whether or not cavitation occurred, and then the probability of cavitation was calculated. In Fig. 3 we show typical results for the probability of nucleation obtained in this way.

The results of this experiment can be analyzed in terms of Eq. 4. It is necessary to first make a quantitative estimate of the experimental volume V and the experimental time τ, allowing for the fact that the maximum negative pressure occurs at an isolated point in space and only once during each cycle of the sound wave. Thus, it is not sufficient to simply take the experimental volume as $(\lambda/2)^3$. We then adjust E_{max} and the prefactor Γ_0 so that the probability $S(T)$ as given by Eq. 4 fits the experimental data as well as possible. The good fit that is obtained demonstrates that that we are indeed seeing a thermally-activated nucleation process.

Our preliminary results from this type of experiment indicate that the nucleation process that is controlling the cavitation has an activation energy that varies from 26 to 33 K as the temperature goes from 0.8 to 1.3 K. Although the activation energy can be determined with reasonable accuracy, the uncertainty in the prefactor is much larger. At present our results indicate that the prefactor is in the range 10^{24} to 10^{27} cm^{-3} sec^{-1}.

One expects that the prefactor should be the attempt frequency ν_{nucl} for nucleation multiplied by the number density n_{nucl} of independent sites at which nucleation can occur. For bulk nucleation n_{nucl} would then be the reciprocal of the volume of the critical nucleus, and for nucleation on a vortex line n_{nucl} should be the length of line per unit volume divided by the linear dimensions of the critical nucleus. This argument gives a prefactor which for nucleation in the bulk is in the range 10^{30} to 10^{31} cm^{-3} sec^{-1}, and for nucleation on vortices is $10^{17} L_{vort}$ cm^{-3} sec^{-1}, where L_{vort} is the length of vortex line in cm per unit cc. The preliminary experimental results for the prefactor are thus about 5 orders of magnitude below the bulk prefactor, and suggest that the dominant nucleation process may in fact

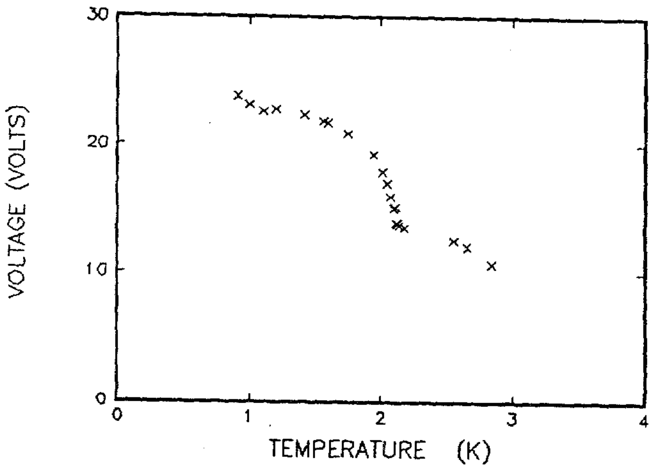

Fig. 4. Transducer excitation voltage required to induce cavitation in liquid helium as a function of temperature.

be the formation of bubbles on vortices. If the nucleation is via vortices a vortex density of 10^7 to 10^{10} cm^{-2} is needed in order for the prefactor to have the right value. This does not appear to be unreasonable given the high sound wave amplitude in the focal region. Milliken *et al*[26] have shown that for a sound intensity smaller than ours by about a factor of 10^6 a vortex density in excess of 10^4 cm^{-2} is created.

Finally, we show in Fig. 4 the results of new measurements of the cavitation strength versus temperature.[30] Because of the uncertainties in the determination of the absolute value of the pressure swing at the acoustic focus we show here a plot of the drive voltage V_{exc} that must be applied to the transducer in order for cavitation to occur. The data were taken while varying the temperature along a series of isopycnals, but we have corrected the results to generate values for V_{exc} as a function of T at the saturated vapor pressure. The temperature dependence of the data is consistent with the earlier measurements of Xiong and Maris[25] in the lower temperature part of the range. In the temperature range near to the lambda point there is a very strong drop in tensile strength, confirming that the decrease suspected by Nissen *et al* is indeed a real effect.[24] Above T_λ the tensile strength continue to decrease, but slowly. The explanation of this strong temperature dependence is not obvious. The tensile strength as governed by nucleation in the bulk is expected to show a small anomaly at T_λ due to the weak singularity that exists in the surface tension. However, the effect that we observe is much larger than would be expected based on this mechanism. If, on the other hand, we suppose that the nucleation occurs on vortices then either the activation energy must be decreasing as $T \rightarrow T_\lambda$, or the amount of vortex line must be undergoing a very rapid increase. A rapid increase in L_{vort} as $T \rightarrow T_\lambda$ has been predicted by Williams.[31] However, his theory is concerned with the equilibrium density of vortex line, whereas in our experiment the density of vortices is probably increased substantially by the sound field. We hope to resolve these issues via forthcoming experiments.

5. ACKNOWLEDGMENTS

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