NUCLEATION OF BUBBLES ON ELECTRONS IN LIQUID HELIUM

HUMPHREY J. MARIS and DENIS KONSTANTINOV
Department of Physics
Brown University
Providence, RI 02912
USA

1. Introduction

Cavitation in liquid helium has been studied for over forty years, beginning with the work of Misener and Hebert [1] and Beams [2]. The interest in helium stemmed from the expectation that because all other elements freeze at higher temperatures, liquid helium should have exceptionally high purity. It was expected that this would greatly reduce the chance of heterogeneous nucleation, and thus enable the study of homogenous nucleation under controlled conditions. However, the early experiments gave values of the cavitation strength that varied significantly from one measurement to the next [3-11], and were in all cases considerably less than the strength that was anticipated theoretically. Most of these experiments were conducted with large volumes of liquid, typically of the order of 1 cm^3, and used liquid that was part of the main bath of a helium cryostat which could easily contain small particles of frozen air. In 1989, Nissen *et al*. [12] used a hemispherical ultrasonic transducer to focus 566 kHz sound into a small volume of liquid helium (volume ~10^{-5} cm^3). Their measurements gave a cavitation strength that increased rapidly as the temperature decreased. At their lowest temperature of 1.6 K, they estimated that vapor bubbles did not appear in the liquid until the pressure reached about – 8 bars. The difference between the results of Nissen *et al*. and the earlier workers is presumably due to the presence of some centers for heterogeneous nucleation in the liquid. Suppose, for example, that the density of these objects is 100 cm^{-3} and that one such object will result in bubble nucleation when the pressure becomes more negative than -0.1 bar. A measurement on a liquid sample of volume 1 cm^3 will then almost certainly give a cavitation strength of 0.1 bars, whereas a measurement on a sample of volume 10^{-5} cm^3 has a high probability of giving a result that is characteristic of the pure liquid.

Since the work of Nissen *et al*., there have been many studies of bubble nucleation in helium at negative pressures [13], and much of this work is summarized in the articles by F. Caupin and S. Balibar in this book. Measurements have been made in both helium-3 [14-18] and helium-4 [14,17,19,20]. In helium-4, the results indicate that at high temperatures, the barrier against bubble nucleation is overcome as a result of thermal fluctuations, whereas below about 0.2 K quantum tunneling through the barrier is the dominant process [14,19,20]. In helium-3, nucleation by quantum tunneling has not yet been clearly demonstrated [16,18]. In these studies, it is believed that bubbles are forming as a result of homogeneous nucleation.

Study of heterogeneous nucleation in most liquids is difficult because it is not usually easy to determine the type and characteristics of the centers responsible for the nucleation. In liquid helium, however, there is one type of "defect", the injected electron, that is ideally suited for a study of heterogeneous nucleation [21]. In this article, we describe studies that have been made of bubble nucleation by electrons and discuss some of the remaining open questions in this field.

2. Electrons in Liquid Helium

At large distances an electron is attracted to a helium atom because the helium atom is polarizable. However, at short distance there is a strong repulsion. This arises because when the electron is within the atom it is forced by the Pauli principle to go into a higher energy state with principal quantum number *n* of 2 or greater. As a result, an electron entering helium has to overcome a potential barrier of approximately 1 eV [22]. If an electron is injected into helium and comes to rest in the liquid, it forces open a spherical cavity which is almost free of helium atoms. The energy of this so called "electron bubble" can be approximated by the expression

$$E = \frac{h^2}{8 m_e R^2} + 4\pi R^2 \alpha + \frac{4\pi}{3} R^3 P, \tag{1}$$

where R is the bubble radius, m_e the electron mass, α the surface tension, and P the pressure. The first term is the zero point energy of the electron confined in a spherical cavity. The second and third terms represent the surface energy of the bubble and the work done to form the bubble against the applied pressure, respectively. For zero pressure and low temperature, the bubble radius R_0 that gives the minimum total energy is $(h^2 / 32\pi m_e \alpha)^{1/4}$. This is 19 Å for helium-4 and 24 Å for helium-3.

In the derivation of Eq. 1 the following assumptions are made:

1) The wave function of the electron is taken to go to zero at the bubble wall, i.e., the potential barrier provided by the helium is assumed to be large.
2) The long range attraction of the electron to the helium atoms is neglected, i.e., the potential energy for the electron inside the bubble is taken to be zero.
3) The width of the bubble wall, i.e., the interface between the liquid and vapor, is assumed to be small compared to the bubble radius.
4) It is assumed that the effect of the helium vapor inside the bubble can be neglected.

Effects 1)-3) are all fairly small corrections [23], and their inclusion is estimated to change the size of the electron bubble by only 5 to 10%. Effect 4) becomes important at temperatures above about 3 K, and is discussed in more detail in the following section.

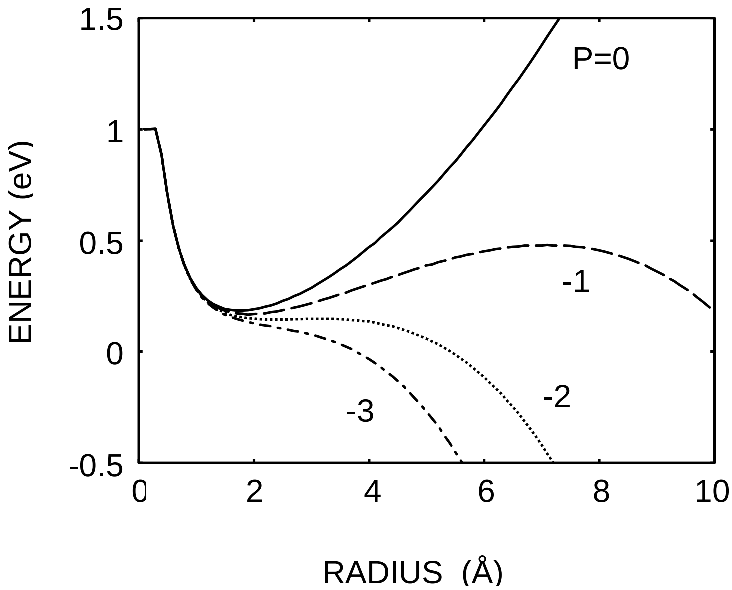

Figure. 1. Energy of an electron bubble as a function of the radius. The different curves are labeled by the pressure in bars.

An electron forms a bubble state because the energy of this state is less than the 1 eV energy the electron would have if it were moving through the uniform liquid. In the absence of applied pressure, the radius of the bubble is $(h^2 / 32\pi\, m_e \alpha)^{1/4}$, and the energy of the electron bubble calculated from Eq. 1 is

$$E_0 = (2\pi\, h^2 \alpha / m_e)^{1/2}, \qquad (2)$$

which for helium-4 is 0.2 eV. It can be seen that the existence of electron bubbles is in large measure a consequence of the extremely small value of the surface tension of helium (0.375 erg cm^{-2}). Since the 1960's these bubbles have been studied by a number of techniques and their basic properties are well established [21].

3. Explosion of Bubbles at Negative Pressure

When a positive pressure is applied to the liquid, the radius at which the energy of the bubble is a minimum decreases. It is easy to show from Eq. 1 that for small pressures the variation of the equilibrium radius with pressure is

$$\frac{dR}{dP} = -\frac{R_0^2}{8\alpha}\ , \tag{3}$$

which for helium-4 is - 1.2 Å bar^{-1}. A negative pressure makes the bubble expand, and at a critical negative pressure P_c, the bubble becomes unstable and begins to grow without limit. As shown in Fig.1, this occurs because when the pressure is negative with respect to P_c, the energy of the bubble decreases monotonically with increasing radius. Then if a sample of liquid helium contains even a single electron bubble, that bubble will grow to a macroscopic size. The fact that at negative pressure an electron bubble will explode was first recognized by Akulichev and Boguslavskii in 1972 [24]. At low temperatures where the presence of helium atoms inside the bubble can be neglected, the critical pressure as calculated from Eq. 1 is

$$P_c = -\frac{16}{5}\left(\frac{2\pi m_e}{5h^2}\right)^{1/4} \alpha^{5/4} \quad . \tag{4}$$

For helium-4, this gives P_c = -1.9 bars [25]. One can consider that the explosion results from the combination of the liquid pressure and the outward pressure exerted by the electron overcoming the effect of the surface tension.

The calculation of the critical pressure at temperatures above 3 K is complicated by the presence of an increasing amount of helium vapor inside the bubble. Detailed calculations of the configuration of the bubble as a function of temperature and applied pressure have been performed by Classen *et al.* [23]. As the pressure is varied, the bubble evolves as follows. When the pressure equals the saturated vapor pressure, the form of the wave function of the electron is only slightly modified from the form that it has at low temperatures. However, the bubble contains a finite number density of helium atoms, primarily in the region close to the bubble wall where the magnitude of the electron wave function is small and hence the interaction between the electron and the atoms is not large. As the pressure is lowered, the density of the helium atoms increases. The interaction between these atoms and the electron results in a decrease in the amplitude of the wave function in the region near to the wall. When the pressure is close to the instability pressure P_c, the electron is confined to a sphere of radius R_e significantly less than the bubble radius R. There is a layer of vapor filling the region between the volume occupied by the electron and the bubble wall. The calculated explosion pressure as a function of temperature is shown in Fig. 2, along with the results of experimental measurements [23]. Similar results and agreement with theory have been obtained for helium-3 [26].

It is important to note that to a very good approximation this cavitation resulting from electrons in helium is a purely “mechanical” process and is only slightly affected by thermal fluctuations or quantum tunneling. This distinguishes it from homogeneous nucleation. Let us elaborate on this point. In the experiments to study cavitation from electrons, sound of frequency in the range 100 kHz to 1 MHz is used to produce a transient negative pressure. Thus, the time scale of the pressure variation is of the order

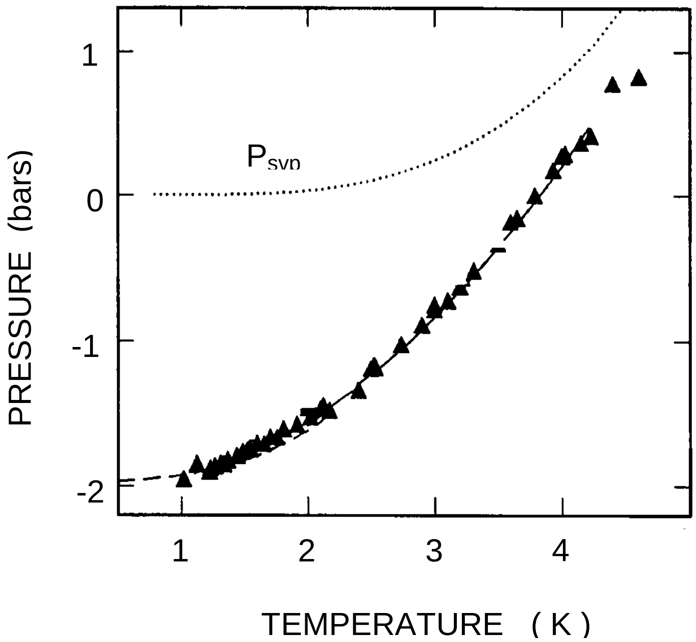

Figure 2. Negative pressure needed to explode an electron bubble in helium-4 as a function of temperature from ref. 23. The solid curve is from theory and the triangles are the measured values. The dotted line indicates the saturated vapor pressure.

of 1 to 10 μs. The frequencies of the normal modes of vibration of the bubble, for example, the breathing mode, are of the order of 10^{10} Hz. As a result, it is correct to consider the sound as producing a quasi-static pressure field which changes very slowly in time and is able to pull the bubble apart. In principle, thermal fluctuations can result in bubble nucleation when the applied pressure P has not yet reached P_c. However, for this to happen, the energy barrier must have decreased to a value that is comparable to kT , e.g., $20kT$. From Eq. 1, it can be shown that for P close to P_c the energy barrier is

$$\Delta E = \frac{32}{15\sqrt{2}} E_0 \left(\frac{P - P_c}{P_c} \right)^{3/2} . \tag{5}$$

Since the energy E_0 in temperature units is 2300 K, this means that $\Delta E >> kT$ unless $(P\text{-}P_c) / P_c$ is extremely small. Thus, thermal fluctuations can help the bubble explode only when P is very close to P_c. A detailed calculation [27] shows that quantum tunneling is also unimportant except very close to P_c.

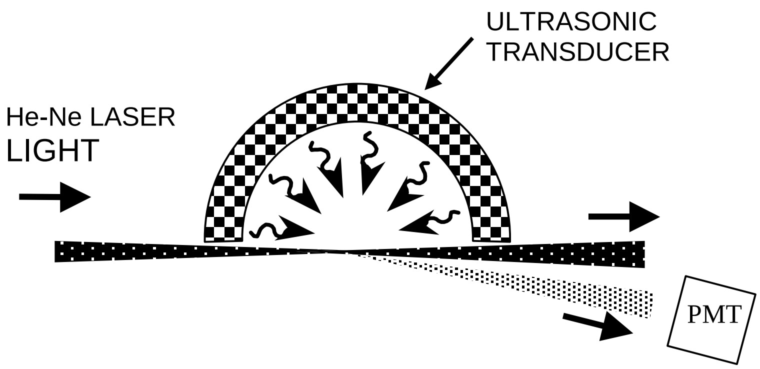

Figure 3. Schematic diagram of the experimental setup to study cavitation in liquid helium. Scattered laser light is detected by the photomultiplier (PMT).

The experimental configuration is shown schematically in Fig. 3. Sound is generated by a hemi-spherical transducer immersed in the liquid, and a large oscillating pressure is produced at the acoustic focus. To a reasonable approximation, the maximum negative pressure produced at the focus is proportional to the driving voltage V_{tran} applied to the transducer. A laser beam is passed through this focus and when a bubble is formed, light is scattered and detected by a photomultiplier. In a typical experiment, the probability that a sound pulse results in cavitation is measured as a function of the driving voltage applied to the transducer. The pressure swing resulting from unit voltage applied to the transducer can be determined through measurements of the cavitation threshold as a function of static pressure in the helium cell [23].

Electrons can be introduced into the liquid either through the use of a radioactive source (β-emitter) or through an electrical discharge at a sharp metal tip in the liquid. Although the radioactive source emits electrons at a constant rate, the number density of electrons in the vicinity of the acoustic focus can be varied by applying a dc voltage to the transducer wall to change the amount of time that each electron spends in the liquid. When a tip is used, the density of the electrons at the focus can again be controlled by varying the applied voltage.

Some typical results are shown in Fig. 4. It can be seen that at a critical transducer voltage V_c of 55 V, the probability of cavitation begins to increase rapidly. At V_c the negative pressure swing at the center of the acoustic focus reaches the magnitude required to explode a bubble. However, the probability of cavitation is still small because nucleation will occur only if there is an electron bubble very close to the center of the acoustic focus. As the transducer voltage is increased, nucleation begins to be possible for electrons further removed from the center of the focus and hence the probability of cavitation increases. From an estimate of the variation of the pressure

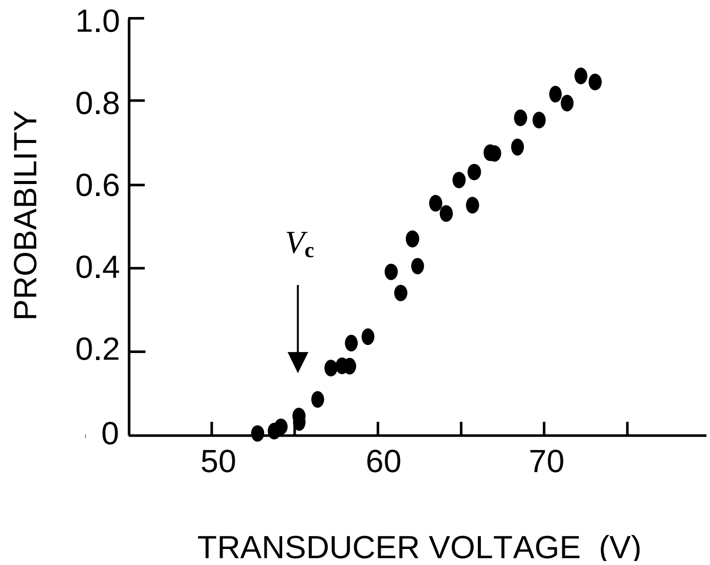

Figure 4. Example of the results from a measurement of the probability of cavitation as a function of applied transducer voltage.

swing with position, it is possible to make a fit to the measured cavitation probability as a function of V_{tran} and from this to deduce the number density of electron bubbles.

4. Some Unsettled Questions

Now we discuss some extra effects that have been seen in the cavitation studies of helium containing electrons.

4.1. Rare Events

In a sample of liquid helium containing electrons, the threshold at which electron bubbles begin to explode is readily identifiable, as was just discussed for Fig. 4. However, it has been found [23] that there may be a small probability of detecting cavitation below this threshold, even for pressure swings much too small to cause the explosion of normal electron bubbles. These rare events are only seen when a radioactive source is used to inject electrons into the helium; they do not appear when electrons are injected from a tip. The probability of these rare events is unaffected by the application of a dc voltage to the transducer, and increases with the duration of the sound pulse. Furthermore, the rare events disappear when the line of sight between the radioactive source and the acoustic focus is blocked.

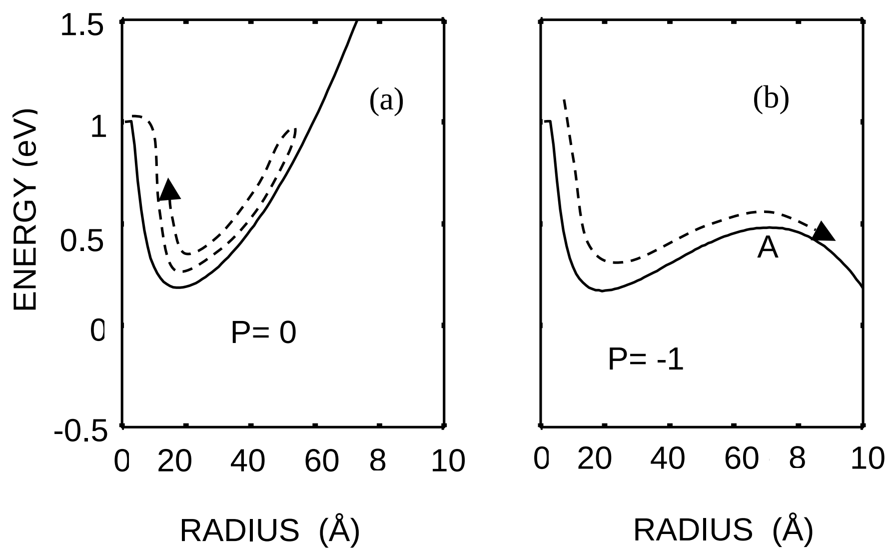

Figure 5. Origin of rare events. See the text for discussion.

These observations suggest that the rare events arise from electrons that pass through the acoustic focus at the same time that the sound oscillation is present. These electrons ionize helium atoms. The secondary electrons then find themselves traveling through liquid that is at a negative pressure. They quickly lose their energy and then begin to form a bubble which grows rapidly. The bubble soon grows to a size such that the energy given by Eq. 1 is a minimum. However, at this point the bubble is surrounded by liquid that is flowing away from it at high velocity. The inertia of this liquid can enable the bubble to keep growing, to overcome the energy barrier, and to continue on to become a bubble of macroscopic size [28].

Figure 5 shows the energetics of this process. When the radius of the bubble is zero, the electron is in uniform liquid helium and has an energy of approximately 1 eV. If the pressure is zero, the bubble will grow to a maximum radius, and then oscillate back and forth until it finally settles at the radius R_0 of minimum energy (Fig. 5a). At a sufficiently large negative pressure, the bubble may continue to grow past the energy barrier (Fig. 5b). How large a negative pressure is required for this to happen? The simplest approach is to ignore any form of dissipation, and to assume that the pressure must be such that the energy at the maximum (point A in Fig. 5b) is less than the starting energy of 1eV. This condition gives good agreement with experiment [28], but is probably a substantial oversimplification. In the first place, account needs to be taken of the compressibility of the liquid. The rapidly expanding bubble will radiate sound into the surrounding liquid, and this will give a loss in energy. A second correction arises from the energy deposited into the liquid *before* the bubble begins to form. Secondary electrons produced by fast particles in helium have an initial kinetic energy of the order of 50 eV and travel a distance of only about 100 Å before losing this energy. Hence, the 50 eV is deposited into a very small volume and there may be a significant local temperature rise. This temperature rise could affect the nucleation process in several ways. For example, the excitations produced in the liquid may further damp the motion of the bubble wall, thereby increasing the magnitude of the negative pressure that is required. On the other hand, an increase in temperature will give a decrease in surface tension and lower the cavitation threshold. The magnitude of these corrections is hard to determine, and has not yet been established.

4.2 Electrons on Vortices

The number of electron bubbles per unit volume of the liquid in these experiments is determined by the balance between the rate at which electrons are injected by the source that is used and the rate at which they escape to the walls of the experimental cell. The escape rate is dependent on the space charge field and the voltage that is applied between the source and the transducer. The electron density also varies rapidly with temperature because of the variation of the bubble mobility with T. In the range 1 to 2 K, the mobility varies with temperature approximately as $\exp(\Delta / T)$ where Δ is the roton energy gap (~8.7 K). As the temperature approaches 1 K, the mobility becomes so large that the number of electrons in the liquid is very small and the probability that cavitation will occur is much less than unity, even when the pressure at the acoustic focus is significantly more negative than the explosion pressure P_c.

However, when the temperature is lowered to around 0.9 K, it is found that cavitation again becomes possible. In this temperature range, the mobility of the electron bubbles is sufficiently large that their velocity exceeds the critical velocity for nucleation of quantized vortices [21]. These are singularities in the liquid that may be in the form of lines ending on the container walls or closed loops entirely within the liquid. The flow of liquid around a vortex is quantized so that

$$\oint \vec{\mathrm{v}}.d\vec{l} = \frac{h}{m_4}, \tag{6}$$

where the integral is along a path enclosing the vortex line, $\vec{\mathrm{v}}$ is the liquid velocity, and m_4 is the mass of a helium atom. When an electron bubble is close to a vortex line, it displaces liquid that has a high velocity and the total energy is reduced. As a consequence, the electron bubble is attracted to the vortex line, and can become bound to it. When electrons become attached to vortices, they move very slowly through the liquid and the electron density then becomes very large.

It is found that when the electron bubbles are attached to vortices the magnitude of the negative pressure at which they explode is reduced by about 12 %. This is shown in Fig. 6. A reduction in the magnitude of the pressure is expected since the circulation of the liquid around the vortex line means that the pressure at the electron bubble will be negative with respect to the pressure in the bulk of the liquid. However, an apparently straightforward calculation [29] of the magnitude of this effect based on a simple model gives a shift of only 4 %, rather than the 12 % found experimentally. The reason for this difference is currently not known. It would be interesting to perform a more sophisticated calculation, using a density-functional method, for example.

4.3 Electrons in Excited States

The discussion so far has been limited to the consideration of bubbles with the electron in the lowest energy quantum state, i.e., the 1S state. Using light of the appropriate wavelength, it is possible to excite the electron to higher energy states. The simplest such transition is from the 1S ground state to the 1P state; at zero applied pressure this transition takes place for a wavelength of approximately 10 μm (energy ~ 0.1 eV). The outward pressure exerted by the electron on the bubble wall is $\hbar^2 \,|\nabla\psi|^2 \,/ 2m_e$; for any state in which the wave function does not have spherical symmetry this pressure varies with direction and hence the shape of the electron bubble that minimizes the total

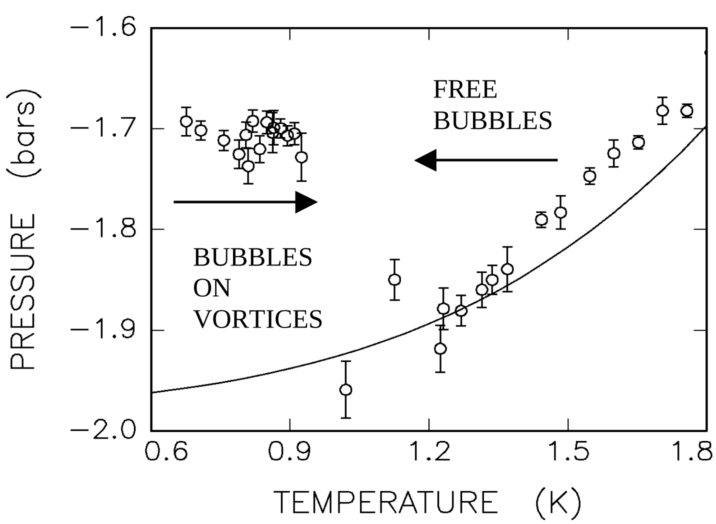

Figure 6. Critical pressure at which electron bubbles explode as a function of temperature from ref. [29]. The solid curve is the theory for a bubble in bulk liquid.

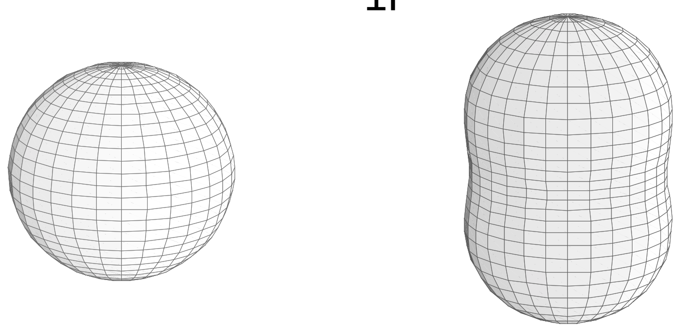

Figure 7. Equilibrium shape of electron bubbles with the electron in the 1S and the 1P states.

energy is no longer spherical. For the 1P state with azimuthal quantum number $m = 0$, the electron pressure is zero in the *x-y* plane, and the bubble has the shape shown in Fig. 7. The pressure at which the electron bubble explodes is different for each quantum state; the magnitude of the negative pressure required to explode the 1P state is calculated to be about 14 % less than for the 1S state [30].

In recent work, a CO_2 laser has been used to excite electron bubbles to the 1P state [31]. Results obtained at 1.99 K are shown in Fig. 8. It can be seen that the effect of the light is to create some new objects that break at a smaller negative pressure. These are the 1P electron bubbles. The transducer voltage required to explode them is about 16 % less than the voltage needed for the 1P bubble, and is thus in good agreement with theory. As expected, the number of these bubbles that are present is proportional to the laser intensity. From measurements of this type we are currently trying to determine an accurate value for the lifetime of the 1P excited state. As a result of re-emission of a photon, the 1P state should have a lifetime of the order of 10 μs [32]; our preliminary measurements indicate a lifetime much shorter than this, presumably due to some form of non-radiative transition.

Finally, it is of interest to consider in more detail what happens when an electron bubble is excited by light. According to the Franck-Condon principle, one should consider that the transition of the electron state takes place before the shape of the bubble changes. After the transition, the pressure on the bubble wall is changed and the wall then begins to move in response to this change in pressure. The motion that ensues is dependent on the magnitude of the dissipative forces acting on the bubble wall. If these are large, the shape of the bubble will change slowly from spherical to the peanut shape shown in Fig. 7. However, if the dissipation is very small, the bubble will

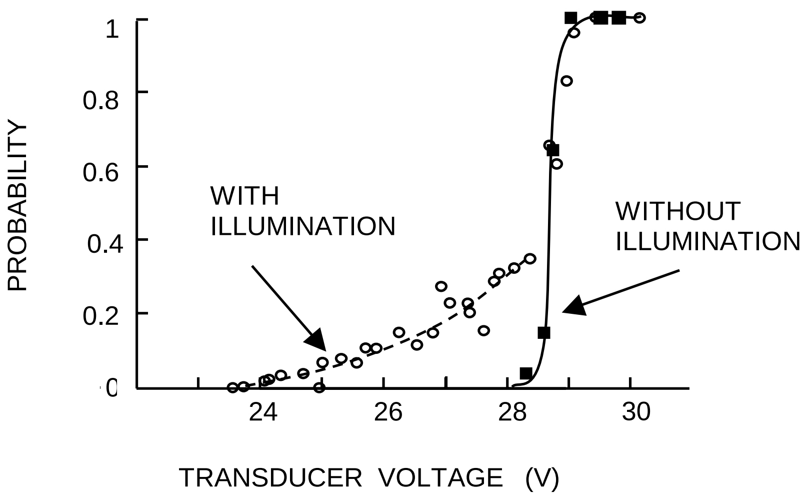

Figure 8. Probability of cavitation with and without illumination. Light produces bubbles containing 1P electrons which are more easily broken.

quickly reach the equilibrium shape, and the inertia of the liquid around the bubble will then keep the bubble wall moving so that the bubble shape becomes even more distorted. At 1.99 K, the temperature at which the data of Fig. 8 were obtained, the dissipation in liquid helium is large and so the bubble should slowly relax to the equilibrium 1P shape. However, as the temperature is lowered below about 1.5 K the dissipation becomes very small. At these temperatures, it is possible [33] that the inertial effects are sufficient to make the radius of the waist of the bubble shrink to zero, thus dividing the bubble into two daughter bubbles. What happens after this is not yet established. Experimental measurements in this temperature range give results that have a complicated dependence on the temperature and the ambient pressure in the experimental cell [31]. For example, when the pressure is zero some 1P bubbles are produced but at 1 bar, where the energy difference between the 1S and the 1P states is better matched to the energy of the photons from the CO_2 laser, no 1P bubbles are detected. Instead, there appears to be a reduction in the density of “normal” electron bubbles, together with the production of a large number of new bubbles that explode only when a larger negative pressure is applied. An additional intriguing result is that at temperatures below 1.5 K, the new bubbles that are produced have a long lifetime, at least one second. Thus these new objects that are produced by the light must be unable to relax back to ordinary electron bubbles by radiative decay. We hope to report on these experiments in more detail in the near future.

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