Detection of Excited-State Electron Bubbles in Superfluid Helium

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We report on experiments in which the pressure oscillation associated with a sound wave is used to explode electron bubbles in liquid helium. Using this technique, we are able to detect the presence of electron bubbles in excited states.

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When an electron is injected into liquid helium, it forces open a cavity from which the helium atoms are excluded [1]. The size of this "electron bubble" is such as to minimize the total energy given by

$$E = E_{el} + A\alpha + PV, \tag{1}$$

where E_{el} is the energy of the electron, A is the surface area of the bubble, α is the surface tension of helium, P is the applied pressure, and V is the volume of the bubble. For a spherical bubble of radius R with the electron in the $1S$ ground state, the energy of the electron is $h^2/8mR^2$, where m is the electron mass. For zero applied pressure, the bubble then has minimum energy when the radius is 19 Å [2]. These bubbles have been studied principally through measurement of ionic mobility [1]. Grimes and Adams [3] and Parshin and Pereversev [6] have used special techniques to make direct measurements of optical absorption. These optical experiments are extremely difficult because the space charge field limits the density of bubbles that can be maintained in the liquid. In this Letter, we report on a new technique which makes it possible to detect bubbles containing electrons in an excited state.

The electron bubble has the interesting property that its equilibrium size and shape are strongly dependent on the state of the electron [4]. The outward pressure exerted by the electron is $\hbar^2|\nabla\psi|^2/2m$, where $\nabla\psi$ is the gradient of the electron wave function at the bubble wall. In mechanical equilibrium, this pressure must be balanced by the surface tension and the external pressure. When the electron is excited from the $1S$ state to a higher state, the pressure exerted by the electron changes and as a result there is a change in the equilibrium shape. At temperatures of around 1.8 K or higher, there is a high density of thermal excitations in superfluid helium and so the damping of the motion of the bubble wall is then fairly large. Consequently, it is expected that, after light is absorbed, the bubble will simply relax to it new equilibrium shape in a time of the order of 20 to 100 ps without significantly overshooting the equilibrium configuration [4]. Recently, the shape corresponding to several different quantum states has been calculated [4,7]. In Fig. 1, we show the shapes of the $1S$ and $1P$ bubbles for three different pressures [8]. In this Letter, we report on the detection of these excited state bubbles using a novel ultrasonic technique.

It can be seen from Fig. 1 that the shape of the $1P$ bubble changes drastically with pressure. For positive pressure, the waist of the bubble shrinks and the shape becomes similar to a peanut. This is because the wave function vanishes in the plane $z = 0$ and so there is no outward pressure exerted by the electron. Thus, around the waist of the bubble, the inward applied pressure has to be exactly balanced by the surface tension. This means that, at the waist of the bubble, the sum of the principal curvatures of the surface has to be negative. For negative pressures, the bubble becomes closer to spherical. It can be shown that, at a critical negative pressure P_c, the bubble becomes unstable and begins to grow without limit. For the $1S$ state, this pressure has been calculated to be -1.89 bars [9]; experiments are in good agreement with this [5]. For the $1P$ state, the calculated value of P_c is -1.63 bars [9]. The existence of the critical pressure P_c provides a new method for the detection of $1P$ bubbles. If a negative pressure is applied to liquid helium containing only $1S$ bubbles, bubbles will be seen to explode only after the pressure passes -1.89 bars. However, if the liquid contains any $1P$ bubbles, these will explode before this pressure is reached. It is important to note that these

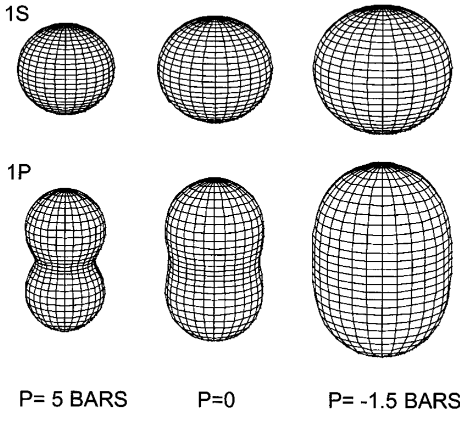

FIG. 1. Equilibrium shapes of electron bubbles in the $1S$ (ground state) and $1P$ state at pressures of -1.5, 0, and 5 bars. The radius of the $1S$ bubble at zero pressure is 19 Å.

explosions are to a very good approximation deterministic events, as distinct from stochastic nucleation processes in which the system has to pass over a barrier as a result of thermal fluctuations or through a barrier by quantum tunneling [10].

The experiment is shown schematically in Fig. 2. Electrons are injected into the liquid from a tungsten tip held at a potential V_0 of -500 to -1000 V. A hemispherical PZT ultrasonic transducer is used to generate 1.4 MHz sound waves in liquid helium. These waves pass through the acoustic focus which has linear dimensions of the order of 10^{-2} cm. The inner wall of the transducer is held at ground potential. Under the influence of the electric field beneath the transducer, the electrons emitted from the tip drift to points on the transducer surface. If an electron bubble is in the vicinity of the acoustic focus and the negative pressure swing is large enough, the bubble will explode and grow quickly. To detect the bubble, a He-Ne laser beam is passed through the acoustic focus and light scattered from the bubble is recorded using a photomultiplier (PMT). The experiment consists of applying a series of 1.4 MHz voltage pulses of amplitude V to the transducer (seven cycles per pulse) and recording the probability S that a bubble is produced. S is defined as the number of sound pulses that lead to cavitation divided by the total number of sound pulses that are applied. The result is shown by the curve labeled "no irradiation by CO_2 laser" in Fig. 3. For the range of transducer voltages used in the experiment, the amplitude of the pressure swing at the focus is approximately proportional to the transducer voltage. If V is below a threshold value V_c then, even at the center of the acoustic focus, the negative pressure never becomes sufficient to explode a $1S$ bubble and so S is zero. For $V > V_c$, a bubble will explode if it is within a certain volume around the acoustic focus. Since the size of this region increases with increasing V, S also increases rapidly with V. From the measured variation of S with V and a calculation of the acoustic field around the focus [5], it is possible to determine that the number density of the electron bubbles is 5.6×10^8 cm^{-3}.

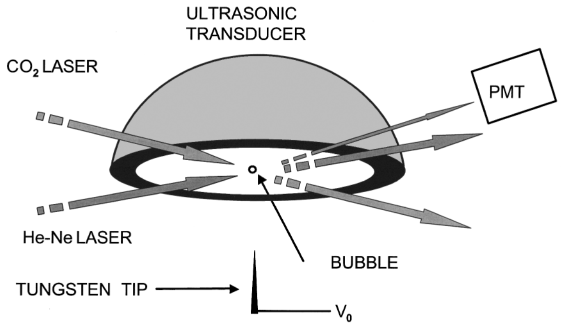

FIG. 2. Schematic diagram of the experimental setup. The beams from the CO_2 and the He-Ne laser both pass through the acoustic focus. Light from the He-Ne laser that is scattered by a bubble is detected by the photomultiplier tube (PMT).

To produce $1P$ electron bubbles, we direct light from a CO_2 laser [11] onto the region containing the acoustic focus. The laser radiation has a photon energy E_ν of 0.1167 eV, and is applied as a pulse starting several hundred μs before the application of the sound and continuing until the sound has passed through the acoustic focus. The photon energy for the $1S \rightarrow 1P$ transition varies as the inverse square of the bubble radius, and, hence, increases with increasing pressure [12]. The measurements of optical absorption by Grimes and Adams [3] indicate that the center of the absorption line matches E_ν when the liquid pressure is around 1 bar and that the line is sufficiently wide that there is still significant absorption at $P = 0$. Results for the cavitation probability in the presence of laser illumination at 1.8 K are included in Fig. 3. In this experiment, we did not attempt to make a precise determination of the light intensity inside the experimental cell, but a rough estimate indicates that 10 W of power at the laser output corresponds to a photon flux in the cell of approximately 3×10^{21} cm^{-2} s^{-1}. The $1P$ bubbles produced by the light begin to break at a transducer voltage that is approximately 18% less than what is needed for $1S$ bubbles. This is in reasonable agreement with the 14% reduction calculated theoretically.

The measured curve of probability versus voltage in the voltage range where only the $1P$ bubbles explode is consistent with a number density of these objects of 1.1×10^7 and 3.4×10^7 cm^{-3} for 2.5 and 10 W, respectively. However, it is important to note that, in fact, the number density has a complex time dependence. The absorption cross section for the $1S \rightarrow 1P$ transition will vary by a large amount during each cycle of the sound pulse [7]. Thus, for example, at a point on the sound cycle where the pressure is -1.5 bar, the transition energy has decreased to $\sim$0.08 eV and so there should be very little optical

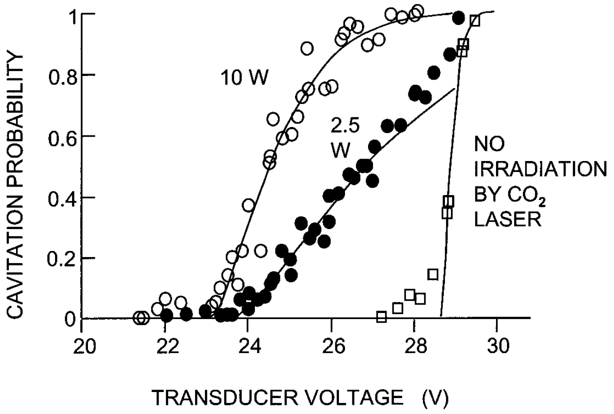

FIG. 3. The probability of bubble nucleation as a function of transducer voltage with and without illumination from the CO_2 laser. Open squares show data obtained with no illumination, solid and open circles are with CO_2 illumination of the indicated power at the laser output. The static pressure in the cell is zero.

absorption. In addition, the $1P$ bubbles have a lifetime due to radiative decay that is approximately 44 μs at $P = 0$ and which varies strongly with pressure [13]. It is possible that the bubbles have a much shorter lifetime due to a nonradiative decay process. By varying the time at which the laser pulse is turned off relative to the time of application of the sound pulse, we have only been able to put an upper bound on the $1P$ bubble lifetime of 40 μs; unfortunately, we are not able to modulate the light from our CO_2 laser on the μs time scale.

Results qualitatively similar to those shown in Fig. 3 are obtained throughout the temperature range 1.8 to 2 K and for pressures 0 to 1 bar. It has been predicted [4] that at lower temperatures the response of the bubble after light absorption will be radically different. When the temperature is lowered, the damping of the bubble wall becomes very small [14]. As a consequence, after the electron makes the transition to the $1P$ state, the bubble will first evolve to a shape similar to the equilibrium $1P$ state already discussed. However, at this point, the liquid around the bubble is still in motion. As a result, the shape will continue to change in a way such that the waist of the bubble may shrink to zero. What might happen after this is not established [14,15]. With the present experimental setup, dramatically different results are indeed seen at lower temperatures (below about 1.4 K), and we briefly mention some aspects of these. The results for the probability have a complex dependence on the duration and timing of the light pulse, and on the pressure. At $P = 0$, the $1P$ bubbles are still seen. At 1 bar, however, no $1P$ bubbles are produced. Instead, the effect of the light appears to be to reduce the apparent number of normal $1S$ bubbles and to produce some new objects that are *harder* to break. Remarkably, these new objects have a long lifetime (> 10 ms). We will make a detailed report on these measurements and discuss possible explanations elsewhere.

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