Journal of Low Temperature Physics, Vol. 134, Nos. 1/2, January 2004 (© 2004)

Experiments to Study the Effect of Light on Electron Bubbles in Liquid Helium

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We give a survey of recent experimental and theoretical work on the effect of light on electron bubbles in liquid helium. The light-induced change in the bubbles is measured using an ultrasonic technique. In helium at temperatures above about 1.7 K, we are able to produce and detect electron bubbles in the 1P quantum state. The properties of the electron bubbles are in agreement with theoretical expectations. However, the application of light to bubbles at low temperatures (T<1.5 K) results in changes in the properties of the bubbles that are not yet understood.

PACS numbers: 67.40.Hf, 43.35.+d

1. INTRODUCTION

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, since 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.[1] 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[2]

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

where E_{el} is the energy of the electron, α is the surface energy of helium, A is the surface area of the bubble, P is the pressure and V is the bubble volume. If the electron is in the ground state, E_{el} equals $h^2/8mR^2$ where m is the mass of the electron, and R is the bubble radius. For zero pressure,

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the bubble radius that gives the minimum total energy is

$$\left(\frac{h^2}{32\pi m\alpha}\right)^{1/4} . \tag{2}$$

This is 19 Å for helium-4 and 24 Å for helium-3.

In the derivation of Eq. 1 it is assumed that the wave function of the electron goes to zero at the bubble wall, i.e., the potential barrier provided by the helium is assumed to be large. The long range attraction of the electron to the helium atoms is neglected, and the potential energy for the electron inside the bubble is taken to be zero. 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. It is assumed that the effect of the helium vapor inside the bubble can be neglected. Inclusion of these effects makes only a small change in the result for the radius of the bubble, at least at low temperatures.[3] In Fig. 1, we show the energy of a bubble as a function of radius as calculated from Eq. 1 for several pressures.

Electron bubbles have been studied principally through measurements of their mobility in an applied electric field. The mobility is limited by the scattering of phonons and rotons from the bubble surface, and hence decreases rapidly as the temperature increases. Optical studies are difficult because it is hard to produce a density of bubbles large enough to give a measurable optical absorption. However, by using special techniques, Grimes and Adams[4] and Parshin and Pereversev[5] have managed to measure the absorption energies required for the electron to make the transition from the 1S ground state to the 1P and 2P excited states. The measured energies are in excellent agreement with calculations based on the simple model that leads to Eq. 1.[6]

Recently, we have considered the process of optical absorption in more detail.[7] When the electron is in a P state, the wave function is no longer spherically symmetric. The electron exerts a pressure on the bubble wall that is large at the poles but vanishes in the equatorial plane. Consequently, the lowest energy shape of the bubble changes and becomes as shown in Fig. 2. This shape changes significantly with pressure; the waist of the bubble becomes very small when the pressure exceeds 5 bars, whereas for negative pressures the bubble becomes closer to spherical. According to the Franck-Condon principle, when light is absorbed the wave function of the electron changes before there is any change in shape of the bubble. If the temperature is high, the damping of the bubble wall by the phonons and rotons in the liquid will be large and the bubble will smoothly relax to the new equilibrium shape.[6] However, at low temperatures the only damping will arise from sound radiation, the bubble will overshoot the equilibrium configuration, and may

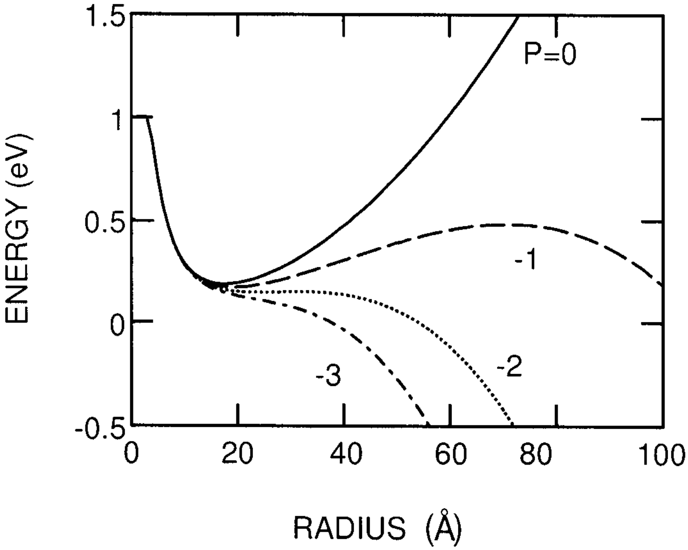

Fig. 1. Energy of an electron bubble as a function of the radius. The different curves are labeled by the pressure in bars.

break into two smaller bubbles. What happens after this is not clear. One possibility is that the two new bubbles are stable and each contain part of the wave function of the electron.[6] A second possibility is that the dynamics of the fission process is such that the two bubbles are formed but only one contains an appreciable amount of wave function.[8] A third possibility is that the interaction of the bubble with the thermal excitations in the helium will cause the quantum state of the system to change in a way such that the final state appears as one normal bubble containing an electron.[9]

Experimental studies of electron bubbles have revealed a number of unexplained phenomena. It has been found that the application of light of the wavelength needed to give the 1S→1P or 1S→2P transitions, results in a change in mobility.[10] The origin of this change is not understood. A second group of experiments has revealed the existence of a family of bubbles of higher mobility than the normal electron bubble.[11] The radius of these bubbles appears to lie between 10 and 15 Å. The nature of these "exotic ions" has not been established but since they have only been seen in a cell in which there was an intense light source, it is natural to wonder if these ions could be related to the fission process just discussed.[6]

2. ELECTRON BUBBLES IN EXCITED STATES

Calculations have been performed of the shape of bubbles in different quantum states.[6,7] In Fig. 2 we have shown the equilibrium shape of the 1S, 1P and 2P bubbles. These shapes are obtained by considering a family of bubble shapes, solving Schrodinger's equation to determine the electron energy inside each shape, and calculating the total energy from Eq. 1. A search is then performed for those shapes for which the total energy increases for any small variation in shape. There seem to be no general theorems concerning this class of problem, i.e., one does not know *a priori* the number of different shapes for which the energy has a local minimum. It was originally assumed that any bubble in which the electron was in an S state would be spherical. Recently, Grinfeld and Kojima[12] have shown that for the nS states with $n \geq 2$, there are multiple non-spherical configurations that correspond to local minima in the total energy. One of these is shown in Fig. 3. As far as we are aware, there has been no corresponding investigation for the P states. This would be very worthwhile but as mentioned above, is not so easy to accomplish because of the absence of any general theorems.

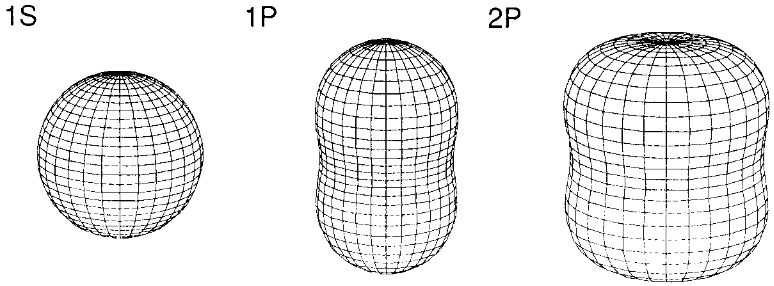

Fig. 2. The shape of electron bubbles in different quantum states in liquid at zero pressure as calculated in ref. 6.

For pressures above about 5 bars, the radius of the waist of the 1P bubble is only a few Å. This is comparable to the width of the liquid-vapor interface and hence calculations based on Eq. 1 cease to be reliable. As a result, it is not known whether there is a stable 1P bubble at higher pressures. For the 2P bubble, a different sort of instability appears at a pressure of 1.53 bars. It can be shown that at this pressure the shape of the bubble becomes such that the 2P and the 1F states are degenerate. The electron can then make a radiationless transition to the 1F state. After this, the energy of the bubble can be lowered continuously by further deformation until the bubble

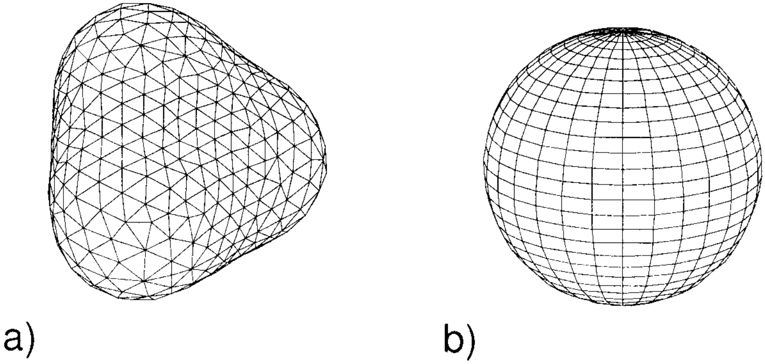

Fig. 3. a) The shape of the 2S state in liquid at pressure −0.75 bars as calculated by Grinfeld and Kojima (ref. 12), compared with b) a spherical bubble with the electron in the 2S state at the same pressure.

breaks into two parts each containing wavefunction of 1P form.[7]

Once the shape of a state is determined, it is straightforward to calculate the cross-section for optical absorption and the lifetime for spontaneous emission.[7] Typical radiative lifetimes to states of lower energy are in the range 1 to 100 μs. The calculated radiative lifetime of the 1P state as a function of pressure is shown in Fig. 4. Note that for all optical transitions there is a large difference between the energy for absorption and emission. For example, at zero pressure the energy needed to excite from the 1S to the 1P is calculated to be 0.106 eV, while the photon emitted in the 1P→1S transition is only 0.041 eV.

It is also interesting to consider bubbles that contain two electrons. Dexter and Fowler[13] showed many years ago that such an object is unstable because its energy is greater than the energy of two single electron bubbles, but they were not able determine whether the two electron bubble was metastable, i.e., stable against small perturbations. Recently, numerical calculations have shown that starting from a spherical two-electron bubble with radius that minimizes the total energy, it is possible to change the shape in a way such that the energy continuously decreases and a final configuration consisting of two single electron bubbles is reached.[7] This calculation *suggests* that stable two-electron bubbles do not exist, but does not prove their non-existence because there is still the possibility of some local minimum in the total energy analogous to the states found by Grinfeld and Kojima.[12]

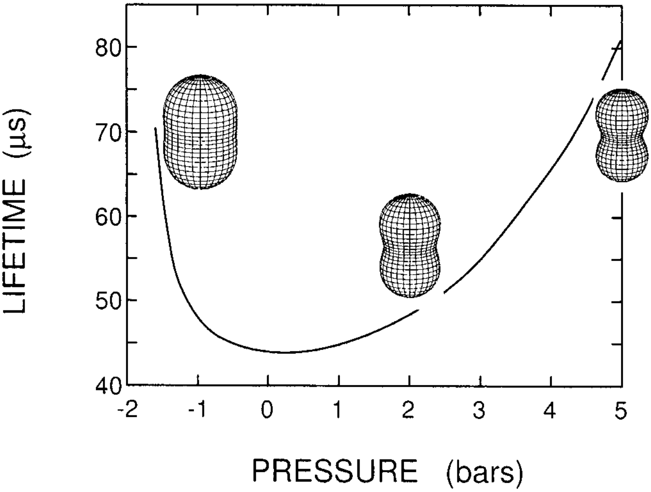

Fig. 4. Radiative lifetime of the 1P state as a function of pressure.

3. STUDIES OF THE 1P STATE

Recently, we have developed a new method for the detection of bubbles in excited states.[14] For each quantum state there is a critical negative pressure P_c at which the bubble becomes unstable against expansion, as shown in Fig. 1.[15] The values of these explosion pressures for different states are listed in Table 1. In the experiment, a hemispherical ultrasonic transducer is used to produce a transient negative pressure in a small region of the liquid. If the pressure becomes more negative than P_c, an electron bubble in the volume will explode and grow to a size large enough to be seen by light scattered from a He-Ne laser. A series of sound pulses is applied and the probability S of detection of an explosion is measured as a function of the transducer voltage V_{tran}. The magnitude of the pressure swing is nearly proportional to V_{tran}. Results are shown in Fig. 5.

Table 1. Explosion pressures for different quantum states.

State	P_c (bars)
1S	-1.89
2S	-1.33
1P	-1.63
2P	-1.22
1D	-1.49

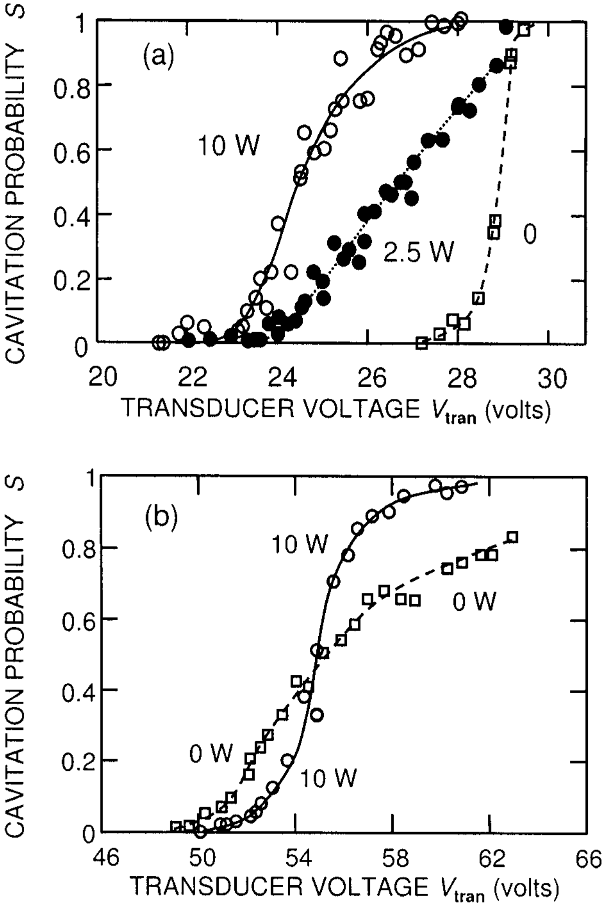

Fig. 5. Probability S of cavitation as a function of the transducer voltage V_{tran} with and without the application of light. a) Measurements at 1.8 K and zero pressure. b) Measurements at 1.45 K and pressure 1 bar. The data are labeled by the laser power in watts and the curves are included as guides to the eye.

Photons from a CO_2 laser have an energy of 0.117 eV, close to the energy required to excite bubbles to the 1P state. In the absence of illumination from the laser, the threshold voltage for explosion of electron bubbles is 27.5 V. At this voltage, ground state electron bubbles begin to explode. It can be seen from Fig. 5a that illumination of the cell with CO_2 laser light results in the production of new objects that break at smaller value of V_{tran}. These new objects are 1P bubbles and the threshold voltage needed to explode them is consistent with the value of P_c listed in Table 1.

From the variation of S with V_{tran}, and the known intensity of the laser light in the cell and absorption cross-section, it is possible to determine the lifetime of the 1P bubbles. Our preliminary estimate of this lifetime is 60 ns[16], considerably less than the radiative lifetime of 40 μs. This indicates that the 1P bubbles relax back to the ground state by some form of non-radiative decay process. There is currently no theory of this process.

At lower temperatures, the changes in the $S - V_{\text{tran}}$ relation induced by illumination becomes qualitatively different. Representative data are shown in Fig. 5b. When the pressure is 1 bar, no bubbles that are easier to break are produced. Instead, illumination reduces S in the voltage range immediately above threshold, but increases S at higher voltage. Thus, the data appear to indicate that the effect of light is to reduce the density of normal electron bubbles and to produce new objects that are harder to break. Remarkably, when the ambient pressure in the cell is lowered to zero, 1P bubbles are produced even though the absorption cross-section for the CO_2 light is much smaller than it is at 1 bar. These results suggest that at 1 bar the bubbles reach the fission condition, but at $P = 0$ they are unable to and so end up in the 1P state. We will discuss these results and their analysis in more detail elsewhere.

ACKNOWLEDGMENTS

We thank S. Balibar, F. Caupin, D.O. Edwards, V. Elser and P. Grinfeld for helpful discussions. This work was supported in part by the National Science Foundation through Grant No. DMR-0305115.

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