Optical Properties of 1P State Electron Bubbles in Liquid Helium-4

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Abstract. When an electron is injected into liquid helium, it forces open a cavity that is free of helium atoms (an electron bubble). If the electron is in the ground 1S state, this bubble is spherical. By optical pumping it is possible to excite a significant fraction of the electron bubbles to the 1P state; the bubbles then lose spherical symmetry. We present calculations of the energies of photons that are needed to excite these 1P bubbles to higher energy states (1D and 2S) and the matrix elements for these transitions. Measurement of these transition energies would provide detailed information about the shape of the 1P bubbles.

An electron injected into liquid helium forces open a small cavity in the liquid referred to as an electron bubble. The energy of this bubble is given by the approximate expression [1]

\[ E = E_{el} + \alpha A + PV , \]

where \( E_{el} \) is the energy of the quantum state of the electron, \( \alpha \) is the surface tension of the liquid, \( A \) is the surface area of the bubble, \( P \) is the pressure, and \( V \) is the volume. The energy \( E_{el} \) depends on the quantum numbers of the state and on the shape and size of the bubble. For a given set of quantum numbers of the electron one can find the lowest energy configuration of the bubble.

In the ground state these electron bubbles have been studied [2] extensively principally through mobility measurements and by investigations of the photon energies at which they absorb light [3]. At a critical negative pressure \( P_c \) of around -1.9 bars, a ground state bubble (1S) becomes unstable against expansion. Measurement [4] of this critical pressure gives good agreement with theoretical calculations [5] and provides another test of the underlying theory.

For a spherical bubble, the quantum states are characterized by a radial quantum number and by angular momentum quantum numbers \( l \) and \( m \). When the electron is raised into an excited state the size and shape of the bubble changes and the lowest energy configuration is, in general, non-spherical.

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For example, the energy of the 1P state with $m = 0$ can be lowered by stretching the bubble along the positive and negative z-axes into a peanut shape. The 1P states with $m = \pm 1$ are mechanically unstable against small perturbations which lack azimuthal symmetry and these states evolve into stable configurations which are identical to the configuration found for the 1P with $m = 0$ state but with an axis of azimuthal symmetry pointing in an arbitrary direction $[6]$. The 1P state has a lifetime $[7]$ of only about 45 ns against decay back to the ground state and so it has not been possible to make mobility measurements for bubbles in this state. The explosion pressure of 1P bubbles has been measured and found to be in reasonable agreement with theory.

![Figure 1. Photon energy required to excite the 1S bubble to the 1P state as a function of pressure. The bubble shapes at -1, 0, and 5 bars are shown.](image)

To produce the 1P bubbles it is necessary to use optical excitation from the ground state. The cross-section for the 1S to 1P transition is of the order of $10^{-14}$ cm$^2$. In Fig.1 we show the calculated value of the energy required to excite an electron from the 1S to the 1P state as a function of the pressure; this uses the size of the bubble as estimated from Eq. 1. Experimental measurements are in good agreement with theory $[3]$. It happens that for pressures in the range between zero and around 2 bars, the photon energy that is required is close to the energy of the photons from a CO$_2$ laser, the strongest light source in the infrared. As a result, it is possible to optically pump at an intensity sufficient to maintain a significant fraction of the electron bubbles in the 1P state even though this state has a short lifetime.

According to the Franck-Condon principle, a light-induced transition of the electron state takes place without change in the shape of the electron bubble. Only after the transition has occurred, will the bubble relax to a new equilibrium configuration. To investigate the equilibrium shape of 1P bubbles in more detail it would be interesting to measure the photon energies which can be absorbed by these objects. This would provide a more detailed test of the theory of the size and shape. Within the electric dipole approximation the states that can be reached starting from 1P are the 1D states with $m = 0$ and $\pm 1$ and the 2S state. The calculated energies for these transitions are shown in Fig. 2.
Figure 2. Photon energies required to excite a 1P electron bubble to the 1D state with $m = 0$, the 1D state with $m = \pm 1$, and the 2S state as a function of pressure.

If the 1S ground state bubbles are excited to the 1P state using light that is polarized along the z-axis, the symmetry axis of the bubbles will initially lie along z. However, it is straightforward to show that because of interactions with phonons and rotons in the liquid within a time of less than 100 ps the direction of the axis of the bubble will become randomized. Consequently, in the calculation of the cross-section for light absorption by 1P bubbles we assume that the orientation of the axis of the bubbles is random. The cross-section for transition from the 1P state to state $i$ averaged over directions is then given by the expression

$$
\sigma = \frac{4\pi e^2}{3c} \left[ |v_{1P,\cdot\cdot}|^2 + |v_{1P,\cdot\cdot}|^2 + |v_{1P,\cdot\cdot}|^2 \right] \delta(E_{1P} + \hbar\omega - E_i),
$$

(2)

where $\omega$ is the light frequency. We have calculated these matrix elements and then obtain the results for $\sigma$ which are shown in Fig. 3. The values shown are obtained by assuming that the width of absorption line is 10% of the photon frequency; this is the width of the absorption line for the 1S to 1P transition. The averaged cross-section for the transition to the 1D state with $m = -1$ is equal to the cross-section for the state with $m=1$.

Figure 3. Optical absorption cross-section for transitions from the 1P state electron bubble to the 1D state with $m = 0$, the 1D state with $m = \pm 1$, and the 2S state as a function of pressure.

For a spherical bubble it is straightforward to show that the ratios between the cross-sections to the 1D, $m=0$, 1D, $m=1$, and 2S states are 1:0.75: 0.31. Because of the deviation of the shape of the 1P bubble from spherical, there is a large change in these ratios. Over most of the pressure range considered, $\sigma$ for the transition to 2S is less than 1% of $\sigma$ for the transition to the 1D, $m=0$ state; at around –1 bar the matrix element to the 2S state changes sign and the cross-section becomes zero. In Fig. 4 we show contour plots of the wave functions (in the $x = 0$ plane) for the different states at zero
pressure. One can see that the wave function of the 2S state is greatly distorted from spherical symmetry whereas the modification of the other three states is less marked.

![Figure 4. Contour plots of the wave functions in the $x=0$ plane for the 1P, 1D $m=0$, 1D $m=\pm 1$, and 2S states when the pressure is zero. The contour lines are equally spaced and the outermost line coincides with the surface of the bubble.](image)

One could measure the optical absorption directly [3] as has been done by Grimes and Adams and by Parshin and Pereverzev for the 1S to 1P transition. It should also be possible to detect the excited state bubbles that are produced through determining the negative pressure at which they explode. We hope to report on measurements of this type in the near future.

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