Spectroscopy of electron bubbles in superfluid ^{4}He and normal liquid ^{3}He

Marc Borrell Martínez.
*Facultat de Física, Universitat de Barcelona, Diagonal 645, 08028 Barcelona, Spain.**
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Using a simple analytical model we have calculated the energy levels and excitation energies of electron bubbles in liquid helium at zero temperature and pressure. We have also calculated the oscillator strengths, cross sections and lifetime for several states. When possible, we have compared our results with experimental data and with the results obtained using more sophisticated models.

I. INTRODUCTION

Electrons can be introduced into liquid helium by field-emission, photo-emission or by radioactive source. When a swift electron enters liquid helium, it loses energy by exciting elementary modes of the liquid, quantized vortices and by ionizing helium atoms. When the electron has lost enough kinetic energy the strong repulsion between the helium atoms and the electron forces the electron to open a small cavity. This fast process takes about 9 ps[1] and produces a bubble of about 20 Å radius essentially void of liquid, where the electron is trapped constituting the so-called electron bubble (e-bubble). This occurs because the e-bubble energy is lower than the energy of the electron moving through uniform bulk liquid because of the Pauli-principle repulsion between an electron and helium atoms is strong, while the attractive polarization interactions is weak. Electron bubbles have been studied theoretical and experimentally for many years[2, 3].

Electron bubbles may move inside the liquid. If they do so in superfluid ^{4}He at velocities above the Landau critical velocity, they can nucleate quantized vortices to which might be eventually attached if the bubble velocity is not very large[4]. At low velocities and low temperatures (T), e-bubbles are spherical. Otherwise, they may deform by thermal fluctuations or by hydrodynamic effects[5].

In this work we study e-bubbles in liquid helium at zero temperature and pressure. Helium remains liquid at $T = 0$ because of the weak He-He interaction and because its small mass favors a large zero point motion, both facts preventing the liquid from solidifying as all other substances do.

The bosonic ^{4}He undergoes a normal-superfluid transition at about $T = 2.17$ K. The fermionic ^{3}He also becomes superfluid below $T \sim 2.7$ mK by pairing off pairs of atoms to build a boson entity. Superfluid ^{3}He is a very complicate system that can be in different superfluid phases. The experiments are usually carried out at temperatures ~ 1 K; at these temperatures ^{3}He is a normal fluid whereas ^{4}He is superfluid. "Zero temperature" should be interpreted here as a temperature not much above several tens mK at which thermal effects are negligible but enough to have ^{3}He in the normal phase and ^{4}He in the superfluid phase.

*Electronic address: mborrema8@alumnes.ub.edu

Experimentally, the absorption spectrum of e-bubbles in liquid ^{4}He has been determined at $T = 1.3$ K from zero up to the solidification pressure (~ 25 atm)[6, 7]. It is dominated by dipole transitions from the $1s$ ground state to the first two p excited states that nearly exhaust the Thomas-Reiche-Kuhn sum rule, with a very small contribution form the $3\,p$ excited state.

II. MODEL AND PHYSICAL CONDITIONS

1. The electron is confined in a spherical cavity of radius R_e.

2. The confining potential is a spherical infinite potential well of radius R_e. Experimentally, the electron has to overcome a barrier of about 1.1 eV to penetrate into the liquid. This energy is fairly higher than the energy of the first energy levels of the e-bubble we are interested in.

3. The temperature and pressure are both zero.

4. We shall use the dipole approximation because the wavelength of the absorbed and emitted photons is much much larger than the diameter of the bubble.

Hypotheses 2 and 4 will be validated by the calculations.

A. Electron energies and wave functions

Firstly, we have analytically solved the Schrödinger equation for one electron in a spherical infinite potential well of radius R_e, determining the electron wave functions and energies. The wave functions are expressed in terms of the spherical Bessel functions $j_l(x)$ and the energies depend on the n-th zero of $j_l(x)$, $Z_{n,l}$[8]. Indeed, writing the electron wave function as $\psi(\mathbf{r}) = \mathcal{R}(r)Y_{l,m}(\Omega)$, the Schrödinger equation for the radial part $\mathcal{R}(r)$ can be written as

$$r^2\frac{d^2\mathcal{R}}{dr^2} + 2r\frac{d\mathcal{R}}{dr} + [\alpha^2 r^2 - l(l+1)]\mathcal{R} = 0\,, \qquad (1)$$

where $\alpha^2 \equiv 2mE/\hbar^2$. Writing $\alpha r \equiv x$, this equation becomes

$$x^2\frac{d^2\mathcal{R}}{dx^2}+2x\frac{d\mathcal{R}}{dx}+[x^2-l(l+1)]\mathcal{R}=0\ , \qquad (2)$$

whose solution, regular at $x=0$, is the spherical Bessel function $j_l(x)$. Imposing $\mathcal{R}(R_e)=0$, this implies that $j_l(\alpha R_e)$ =0 and thus αR_e is a zero of $j_l(x)$. Consequently, we have

$$E_{n,l}=\frac{\hbar^2 Z_{n,l}^2}{2mR_e^2}\ , \qquad (3)$$

where m is the electron mass. Defining $k_0\equiv Z_{1,0}/R_e$, $k_1\equiv Z_{1,1}/R_e$, $k_2\equiv Z_{2,1}/R_e$ and $k_3\equiv Z_{3,1}/R_e$, the wave functions corresponding to the relevant levels for this study are

$$\psi_{1,0,0}(\mathbf{r})=A_0\, j_0(k_0 r)Y_{0,0} \qquad (4)$$

$$\psi_{1,1,m}(\mathbf{r})=A_1\, j_1(k_1 r)Y_{1,m}(\Omega) \qquad (5)$$

$$\psi_{2,1,m}(\mathbf{r})=A_2\, j_2(k_2 r)Y_{2,m}(\Omega) \qquad (6)$$

$$\psi_{3,1,m}(\mathbf{r})=A_3\, j_3(k_3 r)Y_{3,m}(\Omega) \qquad (7)$$

where

$$A_0=\left(\frac{2\pi^2}{R_e^3}\right)^{1/2}$$

$$A_1\equiv\left(\frac{4\cdot Z_{1,1}^4}{R_e^3(2(-1+(Z_{1,1})^2+cos(2Z_{1,1}))+Z_{1,1}sin(Z_{1,1}))}\right)^{1/2}$$

$$A_2\equiv\left(\frac{4Z_{2,1}^4}{R_e^3(2(-1+(Z_{2,1})^2+cos(2Z_{2,1})+Z_{2,1}sin(Z_{2,1}))}\right)^{1/2}$$

$$A_3\equiv\left(\frac{4Z_{3,1}^4}{R_e^3((-1+(Z_{3,1})^2+cos(2Z_{3,1}))+Z_{3,1}sin(2Z_{3,1}))}\right)^{1/2}$$

In the above equations, $Z_{1,0}=\pi$, $Z_{1,1}=4.493$, $Z_{2,l}=7.725$, and $Z_{3,1}$ =10.904[8].

B. Energy and equilibrium radius of the e-bubble

The energy of the e-bubble contains electron, surface and volume terms

$$E=\frac{\hbar^2 Z_{n,l}^2}{2mR^2}+4\pi R^2\sigma+\frac{4}{3}\pi R^3 P\ , \qquad (8)$$

where σ is the surface tension and P the applied pressure. We have obtained the equilibrium radius of the bubble R_e by minimizing the total energy with respect to R when the electron is in the $1s$ state. This yields

$$R_e=\left(\frac{\hbar^2\pi^2}{8\pi m\sigma}\right)^{1/4} \qquad (9)$$

The energy spectrum of the electron confined in the bubble is readily obtained from Eq. (3).

III. RESULTS

A. Absorption energies

Table I collects relevant characteristics of both helium isotopes[9] and the equilibrium radius obtained from Eq. (9). It can be observed that the surface tension is quite different for both isotopes, affecting the equilibrium radius and consequently the electron eigenenergies as shown in Tables II and III.

	R_e (Å)	σ(KÅ^{-2})	$\hbar^2/2m_{He}$ (K Å^2)	ρ_0(Å^{-3})
^{4}He	18.87	0.274	6.0597	0.021836
^{3}He	23.78	0.113	8.0418	0.016347

TABLE I: Characteristic properties of liquid helium and radius of the equilibrium e-bubble.

The electron probability densities corresponding to the ground state and first three p states of the ^{4}He e-bubble are shown in Fig. 1. As imposed by the boundary conditions, they all are zero at the bubble surface.

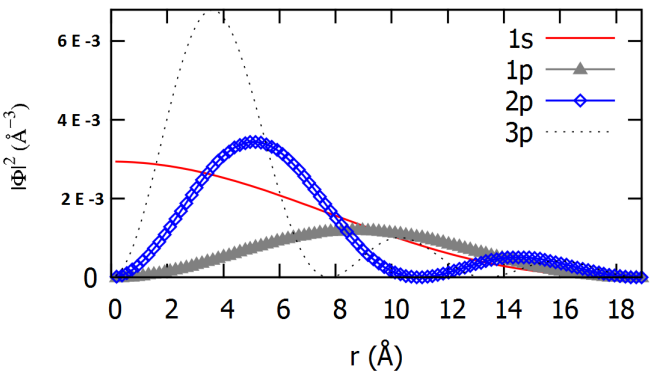

FIG. 1: Electron probability densities for the ^{4}He e-bubble.

The excitation (absorption) energies are obtained from the difference between the energies of the excited and the ground state:

$$\Delta E_{n,l}=E_{n,l}-E_{1,0}=\frac{\hbar^2(Z_{n,l}^2-\pi^2)}{2mR_e^2}\ . \qquad (10)$$

The energies relevant for this study are given in Tables II and III.

n	l	$E_{n,l}$ (eV)	$\Delta E_{n,l}$(eV)
1	0	0,106	-
1	1	0,216	0,110
2	1	0,639	0,533
3	1	1,272	1,167

TABLE II: Electron and absorption energies for the ^{4}He e-bubble.

n	l	$E_{n,l}$ (eV)	$\Delta E_{n,l}$ (eV)
1	0	0,067	-
1	1	0,136	0,070
2	1	0,402	0,336
3	1	0,801	0,735

TABLE III: Electron and absorption energies for the ^{3}He e-bubble.

There are experimental data for ^{4}He but not for ^{3}He e-bubbles[6, 7]. Although they have been obtained at $T = 1.3$ K, the surface tension is not much different from that at zero temperature and can be compared with the results of our model. The experimental $1s \to 1p$ absorption peak is at ~ 0.103 eV (see Fig. 2). This value is very close to ours (0.106 eV) and indicates that the infinite potential well model works well. More fundamental approaches that in particular incorporate the finite height of the confining potential and that it goes smoothly instead of abruptly to zero yield 0.104 eV [10, 11] The experimental $1s \to 2p$ absorption peak is at ~ 0.49 eV whereas we have obtained 0.53 eV. There is no experimental evidence for the transition to the $3p$ state likely because it carries very little strength, see below. Our model yields an energy of 1.167 eV for the $1s \to 3p$ transition to be compared with the value of 0.936 eV obtained in the mentioned reference[10].

We conclude that the differences between our energies and those obtained with more realistic methods and models, as well as with the experimental results, basically arise from the infinite potential well we have used that has allowed us to carry out the calculations analytically. Notice that the energy of the $3p$ state is similar or even larger than the experimental value of the confining potential well, so one should not expect that our model works for this and higher energy states.

B. Oscillator strengths

The oscillator strength is a dimensionless quantity that expresses the probability of absorption or emission of electromagnetic radiation in transitions between energy levels in the dipole approximation. It fulfills a model-independent sum rule that may be used to determine how many states are sensibly excited (or de-excited) from a given state, in our case the ground state of the e-bubble. The oscillator strength $f_{f,i}$ is defined as:[12, 13]

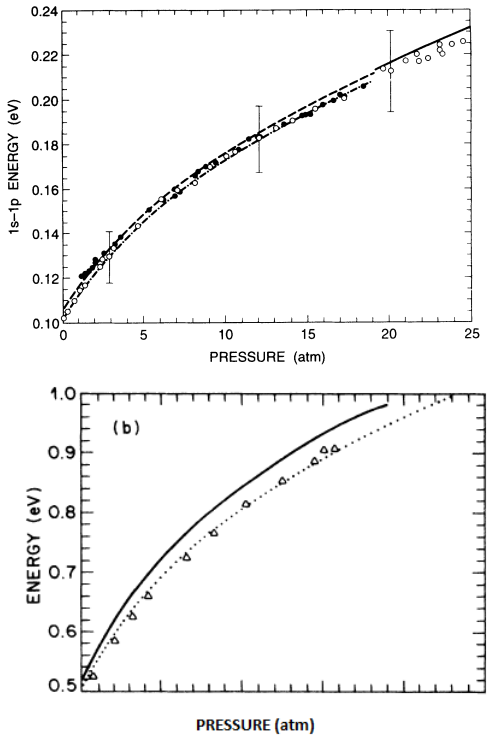

FIG. 2: *E*xperimental absorption energies for the $1s \to 1p$ transition (top) and $1s \to 2p$ (bottom) at $T = 1.3$ K as a function of pressure for the 4He e-bubble[6, 7] (symbols). Also shown is the result obtained from different calculations (lines).

$$f_{f,i} \equiv \frac{2m\omega_{f,i}}{3\hbar}|\langle f|\mathbf{r}|i\rangle|^2 \,, \tag{11}$$

where f and i represent the final and initial states, respectively, $\omega_{f,i}$ is the transition frequency $(E_f - E_i)/\hbar$ and m is the electron mass. The matrix element entering the definition of the oscillatory strength embodies the selection rules for an electric dipole transition, namely

$$\Delta l = \pm 1; \Delta m = 0, \pm 1; \Delta m_s = 0 \,.$$

It is for this reason that we have limited the calculation of the excited states to np states.

The minimum wavelength of the absorbed photons (maximum energy) corresponds to the transition $1s \to 3p$ for ^4He (1.167 eV) and this wavelength is about $10^4 \mathring{A}$. In comparison with the bubble radius ($\sim 20\mathring{A}$) the wavelength is much higher. Hence, the electric dipole approximation (II.4) is perfectly valid.

It can be shown that[12, 13]

$$\sum_f f_{f,i} = N_e = 1 \,.$$

This expression is the so-called Thomas-Reiche-Khun sum rule. The results of the calculation are given inTable IV.

The sum of the first three oscillator strengths is 0.9969, indicating that the corresponding absorption lines nearly

Transition	Oscillator strength	σ_0(eV cm^2)
1s $\rightarrow$ 1p	0.9667	1.06 E-16
1s $\rightarrow$ 2p	0.0254	2.80 E-18
1s $\rightarrow$ 3p	0.0048	5.30 E-19

TABLE IV: Oscillator strengths and σ_0 values for absorption transitions in e-bubbles.

exhaust the spectrum. This is in agreement with experiments, where only two absorption lines have been detected[6, 7].

The results displayed in Table IV are independent of the isotope. This remarkable result is exact for the infinite potential well of our model, but is also very well fulfilled by other more realistic approaches[11]. Notice that the matrix element $|\langle f|\mathbf{r}|i\rangle|$ is proportional to R_e and the transition energies are proportional to $1/R_e^2$. Consequently, the dependence on the radius of the e-bubble –different for each isotope just because of their different surface tension– cancels out. The same happens for σ_0, whose definition is given below.

C. Total absorption cross section

Within the dipole approximation, the total photoabsorption cross section can be easily obtained. Considering that the electric field is polarized in the x direction, it is given by[12]

$$\sigma = 4\pi^2\alpha\,\omega_{f,i}|\langle f|x|i\rangle|^2\delta(E_f - E_i - \hbar w) \equiv \sigma_0\delta(\hbar\omega_{f,i} - \hbar w)\ , \tag{12}$$

where $\alpha = e^2/(\hbar c)$ is the fine structure constant. Notice that in the dipole approximation the absorption cross section between two discrete levels is a delta function that is smoothed out when one considers that the levels have some width. The value of the σ_0 coefficient is given in Table IV.

As before, our σ_0 values are similar to those obtained in the more sophisticated calculations[10] for the first two transitions. For the third transition we obtain 5.30 $\times 10^{-19}$ eV cm^2 to be compared with the value of 3.25 $\times 10^{-19}$ eV cm^2 obtained in that reference. The origin of the difference is again the simple confining potential we have employed here.

D. Spontaneous emission from an excited p state

To determine the absorption energies we have supposed that the transition from the ground to the np excited state is instantaneous, meaning that the bubble does not change of size nor shape during the excitation, which is justified. A different situation occurs in the de-excitation of the p states (emission spectrum). Experimentally, the lifetime of these states is in the ns timescale[14]. This is a long period of time for liquid helium, that has enough time to relax around the excited electron orbital before it decays by spontaneous photon emission. This can be inforred as follows. The surface modes of a spherical and incompressible helium bubble are[15]:

$$\hbar\omega_\lambda = \hbar\sqrt{\frac{\sigma(\lambda-1)(\lambda+1)(\lambda+2)}{m_{He}\rho_0 R_e^3}}\ , \tag{13}$$

where ρ_0 is the atomic density of the fluid. For the lowest lying quadrupole ($\lambda = 2$) mode, the above equation yields around 0.5 K for the ^{4}He bubble and 0.3 K for ^{3}He. This implies a characteristic time scale (time period of the quadrupole oscillation) of $\sim$ 90 ps for ^{4}He and of $\sim$ 150 ps for ^{3}He, much shorter than the lifetime of the excited state.

The relaxation of the e-bubble around an excited state is illustrated in Fig. 3 for the case of the 1p state[15]. Notice that a large part of the absorbed energy, about 2/3, is transferred to the liquid and only $\sim$ 1/3 of this energy is taken away by the emitted photon.

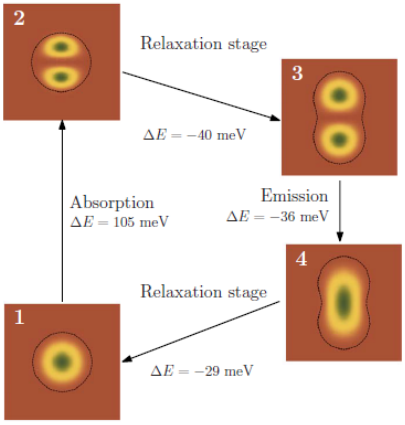

FIG. 3: ^{4}He e-bubble configurations around a 1p excited state at $T = 0$ and zero pressure[15].

Clearly, these relaxation processes and photon emission are beyond the scope of this work. Yet, it might be interested to determine the spontaneous transition rate per unit time of the 1$p \rightarrow$ 1s de-excitation to have a very rough theoretical estimate of the lifetime of the 1p state, having of course in mind the obvious limitations of keeping spherical and unrelaxed the excited e-bubble during the photon emission.

The transition rate per unit time for spontaneous transition from a state i to a state f is[13]:

$$W^s_{f,i} = \frac{2\alpha\,(\Delta E_{f,i})^2}{mc^2\hbar}|f_{f,i}|\ , \tag{14}$$

where the ingredients entering this expression have been previously obtained. The lifetime of the 1p state is

$$\tau_{1p} = \frac{1}{W^s_{1s,1p}} \tag{15}$$

Using this equation we have obtained, for ^{4}He, a lifetime of ~ 2 μs. For ^{3}He the lifetime is ~ 5 μs. More sophisticated calculations give a lifetime of ~ 44 μs[16]. This simple, rough estimate, indicates that the lower energy excited states of e-bubbles in liquid helium at low temperatures and pressures do relax before de-exciting by photon emission. Experimentally this lifetime is estimated to be ~ 50 ns[14]. The origin of this large discrepancy is still unknown.

IV. SUMMARY AND CONCLUSIONS

- We have shown that the energies of the $1s \rightarrow 1p$ and $1s \rightarrow 2p$ excitations of e-bubbles in liquid ^{4}He at zero temperature and pressure can be calculated with a simple analytical model whose basic ingredient is a spherical infinite potential well whose radius is determined by energy minimization. The calculated energies are in good agreement with the experiments and with those obtained by other far more sophisticated models.

- We have shown that the first three dipole excitations nearly exhaust the Thomas-Reiche-Kuhn sum rule, with a very small contribution of the $\rightarrow 3p$ excitation that is not detected in the experiments.

- We have carried out similar calculations for ^{3}He e-bubbles for which there are no experimental results. The differences between the results obtained for both isotopes are essentially due to the different surface tension of the liquids. Remarkably, within our model the oscillator strengths and total absorption cross sections are independent of the isotope. We have check in the literature that this result sensibly holds when the e-bubble is described using more sophisticated methods.

- We have presented arguments indicating that the radiative de-excitation of the e-bubble takes place after it has relaxed around the excited electron. We have carried out very approximate calculations of the lifetime of the excited $1p$ state obtaining approximately the same order of magnitude for it than detailed calculations that take into account the deformation of the relaxed e-bubble. The experiments yield a much shorter lifetime for the $1p$ state. Yet, this time turns out to be much larger than the typical relaxation time of liquid helium around the excited e-bubble.

Lastly, we would like to emphasize the simplicity of the electron bubble as a textbook example of a system whose quantum mechanical description, only comparable in its simplicity to that of the hydrogen atom, allows one to straightforwardly calculate some properties that can be compared with the results of complex experiments in condensed matter and low temperature physics.

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