Instability of the 2*S* Electron Bubbles

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The 2*S* electron bubble placed in liquid helium has been previously believed to be spherical. We show that the 2*S* bubble is morphologically unstable at pressures above −1.23 bars. The 2*S* state being known to be radially unstable at pressures below −1.33 bars, the result leaves only a very narrow pressure range in which it can be found in a spherical configuration. Our stability analysis indicates that the 2*S* bubble is unstable against perturbations proportional to any of the third spherical harmonics Y_{3m}. Our numerical simulations show that there exist nonspherical stable configurations, such as the ones Maris and Konstantinov predicted for the 1*P*, 1*D*, and 2*P* electron bubbles and confirmed experimentally for the 1*P*. We believe that the 2*S* bubbles can also be produced and that our prediction will yield itself to experimental verification.

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Introduction.—Electron bubbles are formed by excess electrons in liquid helium. The helium atoms are pushed away by quantum kinetic pressure exerted by the trapped electron. Quantum kinetic pressure is balanced by surface tension and hydrostatic pressure. Our primary focus is the equilibrium and stability of the *S* electron bubbles under the influence of these three factors.

The equilibrium 1*S* electron bubble shape is a sphere whose radius is theoretically estimated at 19.4 Å. When the hydrostatic pressure P drops below $P_c = -1.88$ bars, the bubble becomes radially unstable and explodes [1,2]. Maris and Balibar call this particular value of pressure "a landmark in the quantum sea" [3]. It is a critical tool for ascertaining the attained pressure in experiments on homogeneous cavitation in liquid helium in which negative pressures must be produced far away from the walls of the container and cannot be measured by conventional means.

Higher state electron bubbles explode at higher pressures. The critical pressure for the 1*P* bubbles has been calculated to be −1.69 bars, and for the 2*S* it is −1.33 bars [4,5]. The prediction for the 1*P* bubbles has been recently confirmed experimentally by Konstantinov and Maris [6].

Stability analysis of electron bubbles until now has focused on radial perturbations. We study morphological stability. This is necessary to conclude the overall stability: A system is unstable if there is even a single infinitesimal perturbation that diminishes its total energy. It is quite common for a system to be radially stable but morphologically unstable [7]. Electron bubbles are relatively simple systems (compared, for example, to multielectron bubbles whose stability properties are also an active area of research [8]), and we feel that their stability properties ought to be well understood. Our key conclusion is that the 2*S* electron bubbles are morphologically unstable at pressures $P > -1.23$ bars. We present an analytical proof of this effect and we are still searching for a simple physical explanation. The result does not affect the "landmark" pressure since the 1*S* electron bubbles are stable against all infinitesimally small perturbations for $P > P_c$.

We consider the stability problem in the framework of the theoretical model presented in [1]. In that Letter, the authors postulate the total energy E and analyze the radial stability of the 1*S* electron bubble with respect to radial perturbations. In its simplest form, the model includes quantum kinetic pressure, surface tension, and external pressure:

$$E = E_{el} + \sigma A + VP, \tag{1}$$

where E_{el} is the energy of the electron, σ is the surface energy density of the helium/void interface, A is the surface area of the bubble, and V is its volume. We do not include the electrostatic energy. Experiments have shown that the simple model, which ignores the electrostatic effects, provides accurate predictions for observations such as the pressure at which electron bubbles explode.

The energy of a spherical 1*S* bubble of radius R can be written as

$$E = \frac{h^2}{8mR^2} + 4\pi\sigma R^2 + \frac{4}{3}\pi R^3 P. \tag{2}$$

At positive pressures P, there is a unique equilibrium radius R_P. At $P = 0$, we have

$$R_0 = \sqrt[4]{\frac{h^2}{32\pi m\sigma}} \approx 19.4 \text{ Å}. \tag{3}$$

At negative pressures, an equilibrium exists (and, in fact, there are two equilibria) only if P exceeds the landmark pressure

$$P_c = -\frac{16}{5}\sqrt[4]{\frac{2\pi m\sigma^5}{5h^2}}. \quad (4)$$

The smaller of the two equilibrium bubbles is radially stable and the larger is unstable.

Our stability analysis is based on the second energy variation. We conclude that the spherical 2*S* electron bubble is unstable against perturbations proportional to the third spherical harmonic Y_{3m} at $P > -1.23$ bars. Further, we conclude that the 1*S* electron bubble is morphologically stable at all pressures (but radially unstable for $P < P_c$). This is consistent with earlier works [9,10], although our approach does not require modeling the dynamics of the surrounding liquid. We perform a series of numerical simulations that reveal nonspherical equilibrium shapes. For the 1*S*, 1*P*, and 1*D* bubbles, our numerical results are in close agreement with simulations performed by Konstantinov and Maris [4], who used a different technique.

Equilibrium equations.—For an arbitrary shape, the equilibrium equation is a balance of pressures:

$$\frac{h^2}{8\pi^2 m}|\nabla\psi|^2 + \sigma\kappa = P, \quad (5)$$

where ψ is the wave function of the electron and κ is the mean curvature of the boundary *S*. This equation is deeply nonlinear since the location of the boundary is unknown. Nonlinearity is a property shared by virtually all problems with unknown boundaries. One of the most vivid examples of this is the problem of the equilibrium shape of a crystalline substance. The reader can find a discussion of this problem in a series of lectures by Nozières [11]. For a spherical configuration, the equilibrium equation becomes ($\kappa = -2/R_p$)

$$\frac{n^2 h^2}{16\pi m R_P^5} - \frac{2\sigma}{R_P} = P, \quad (6)$$

where n is the number of the excited *S* state.

We introduce dimensionless quantities y and a:

$$y = \frac{R_0}{R_p} = \sqrt{\frac{h^2}{32\pi m\sigma R_p^4}}, \qquad a = \frac{PR_0}{2\sigma}. \quad (7)$$

The value of y measures the relative strengths of quantum kinetic and capillary pressures. The dimensionless equilibrium equation for a spherical configuration becomes

$$n^2 y^5 - y - a = 0. \quad (8)$$

Figure 1 contains the graph of $y^5 - y - a$ for four values of a: -1, $-1/4$, 0, and 1. The zeros correspond to equilibrium spherical bubbles. At non-negative pressures ($a > 0$), there is a single equilibrium radius. At negative pressures ($a < 0$), there are two equilibrium radii for $P > P_c$ (smaller—stable; larger—unstable) and none for $P < P_c$.

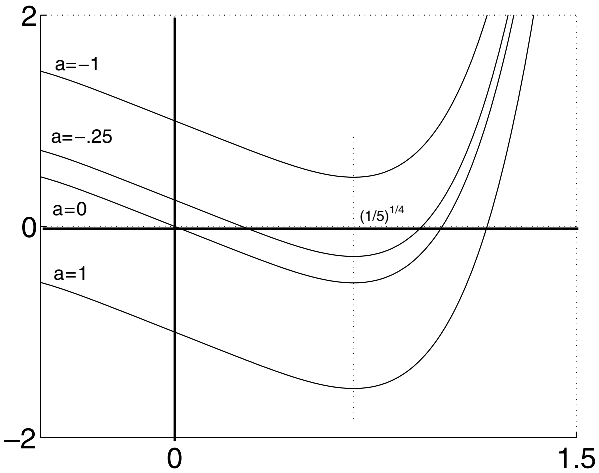

FIG. 1. Graph of $y^5 - y - a$ vs y for several values of a. The minimum occurs at $(1/5)^{1/4}$.

As a historical note, Migdal [12,13] studied the spectrum of an electron trapped in a slightly ellipsoidal cavity and was the first to analyze the perturbation of the spectrum of the Schrödinger operator induced by the deformation of the boundary. His approach can be applied to the question of stability of electron bubbles. An ellipsoidal perturbation is essentially proportional to the second spherical harmonic $Y_{2,0}(\theta, \phi)$ and would miss our instability which is revealed by the third harmonic.

The instability criteria.—The instability criteria are obtained by computing the second energy variation $\delta^2 E$ with respect to infinitesimal perturbations of the boundary *C*:

$$\delta^2 E = -\int_S dSC\left\{\begin{matrix} \sigma(\Delta_S C + CB^\alpha_\beta B^\beta_\alpha) \\ + \frac{h^2}{8\pi m}\nabla\psi(\nabla\partial\psi + CB^\alpha_\alpha\nabla\psi) \end{matrix}\right\}, \quad (9)$$

where $\Delta_S C$ is the surface Laplacian of C, $B^\alpha_\beta B^\beta_\alpha$ is the trace of the third groundform $B^\alpha_\beta B^\beta_\gamma$, and $\partial\psi$ is the perturbation of the wave function ψ induced by the perturbation of the boundary C. For a spherical configuration of radius R_p, $B^\alpha_\beta B^\beta_\alpha$ equals $2R_P^{-2}$ [14]. Equation (9) applies to equilibrium configurations of arbitrary shapes. For spherical equilibrium configurations, analysis can be carried further by representing C as a series in spherical harmonics $Y_{lm}(\theta, \phi)$:

$$C = R_P \sum_{l,m} C_{lm} Y_{lm}(\theta, \phi), \quad (10)$$

where the factor R_P is introduced to make the coefficients C_{lm} dimensionless. Radial perturbations correspond to the zeroth harmonic $Y_{00}(\theta, \phi) = (4\pi)^{-1/2}$. The three $l = 1$ harmonics $Y_{1m}(\theta, \phi)$, $m = -1, 0, 1$, are responsible for the motion of the cavity as a rigid body. The five $l = 2$ harmonics, $Y_{2m}(\theta, \phi)$, $m = -2, -1, 0, 1, 2$, describe an infinitesimal stretching of a sphere into an ellipsoid. The $l > 2$ harmonics represent successively more complicated perturbations of the bubbles's boundary. More detailed derivations of (9) can be found in [15].

When we substitute the expansion (10) into the second energy variation, it becomes a diagonal quadratic form in terms of the C_{lm}:

$$\delta^2 E = \sum_{l,m} A_{lm} |C_{lm}|^2, \tag{11}$$

where

$$\frac{A_{00}}{\sigma R_p^2} = 8\pi(5n^2y^4 - 1),$$
$$\frac{A_{l\neq 0,m}}{\sigma R_p^2} = 4y^4n^2\left(\frac{n\pi J_{l-(1/2)}(n\pi)}{J_{l+(1/2)}(n\pi)} - (l-1)\right) + (l-1)(l+2). \tag{12}$$

$J_k(r)$ is the Bessel function of the first kind. The physical system is stable if the condition

$$A_{lm} \geq 0 \tag{13}$$

holds for every harmonic.

Stability analysis: radial perturbations.—According to Eqs. (12) and (13), the radial stability condition is

$$5n^2y^4 - 1 > 0.$$

For the range of pressures $n^{-1/2}P_c < P < 0$, at which two equilibrium bubbles exist, this condition is satisfied by the smaller radius, which corresponds to the greater positive root of (8), and is violated by the larger radius. At positive pressures, this criterion is always satisfied. These conclusions agree with radial analysis of Eq. (1).

Stability analysis: morphological perturbations.—The electron bubble is morphologically stable if for all $l > 0$

$$4y^4n^2\left(\frac{n\pi J_{l-(1/2)}(n\pi)}{J_{l+(1/2)}(n\pi)} - (l-1)\right) + (l-1)(l+2) > 0. \tag{14}$$

The $l = 1$ harmonic is responsible for the displacement of the cavity as a rigid body. Since $J_{1/2}(n\pi) = 0$, we have $A_{1m} = 0$ which indicates neutral stability with respect to motion as a rigid body. This is to be expected since the physical system is invariant under such motion.

We now turn to the higher harmonics, $l > 1$. For the $1S$ electrons ($n = 1$), it can be shown that, for all $l > 1$,

$$\frac{\pi J_{l-(1/2)}(\pi)}{J_{l+(1/2)}(\pi)} - (l-1) > 0. \tag{15}$$

We therefore conclude that the $1S$ electron bubble is stable with respect to all morphological perturbations.

Instability of the $2S$ electron bubbles.—The spherical $2S$ electron bubble is unstable against $Y_{3m}(\theta, \phi)$ for $P > -1.23$ bars. The numerical form of the stability criterion (14) for $l = 3$ is $y < 0.55$. This value of y (7) corresponds to the equilibrium radius of $R_P = 1.82R_0$, which occurs at the aforementioned pressure of -1.23 bars. Recall that the $2S$ electron bubble is radially unstable for $P < -1.33$. This leaves only a very narrow range of pressures $-1.33 < P < -1.23$ at which the spherical $2S$ electron bubble is stable. Our numerical simulations show that for some pressures above -1.23 bars there exist nonspherical stable configurations. We therefore do not expect that the morphological instability reported here will necessarily result in an explosion characteristic of radial instabilities.

Nonspherical equilibrium shapes.—Our numerical simulations are based on a gradient descent scheme: The surface of the bubble is evolved in such a way that the total energy decreases at every step. The eigenvalue problem is solved by finite elements with tetrahedral elements and linear trial functions. In fact, we adopted a finite element philosophy for the entire problem. We replace the smooth surface of the bubble with a polyhedron and solve the new approximate problem exactly. The surface tension and pressure contributions to the total energy are easily computed for a polyhedron in closed form. The quantum energy of electron is replaced by the finite element estimate for the eigenvalue of the Schrödinger operator. Therefore the computation is sensitive to the interior mesh as well as the exterior polyhedron.

Figure 2 shows the equilibrium spherical configuration (a) at zero pressure (which is unstable) and the equilibrium stable nonspherical configuration (b) at -0.75 bar. The latter has tetrahedral symmetry and is the only stable configuration that we could find at that pressure. It is an open question whether other stable configurations exist. At higher pressures, we observed that eigenstates cross in the course of evolution and the $2S$ state relaxes to the same shape as the $1D$ state reported in [4].

Experimental verification.—How might the predicted tetrahedral equilibrium shape be verified experimentally? In their study [6] of electron bubble "explosion," electrons were excited up to the $1P$ state by a carbon dioxide laser providing a 11 μm wavelength radiation. A similar method might be used. The required energy to excite the electron in the bubble from its ground state to the $2S$ state is 0.3 eV which corresponds to 4.1 μm wavelength. The nearest excited states are at least 0.05 eV away. A tunable solid state laser might be used for the radiation source. Another method would be to pass the radiation from an appropriate blackbody source through a monochromator. To verify that $2S$ states are indeed excited, the negative pressure at which the bubble explodes should

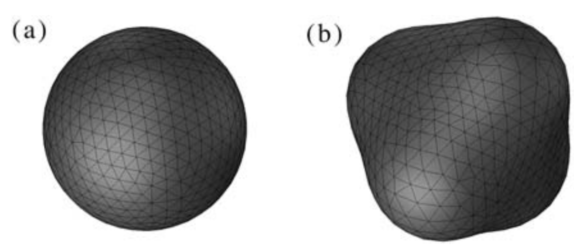

FIG. 2. The $2S$ electron bubble: An unstable equilibrium spherical configuration at zero pressure and a stable nonspherical configuration at -0.75 bar.

be checked to see that it coincides with −1.33 bars. To see that the equilibrium shape is nonspherical, we suggest looking for changes in the mobility of the electron bubbles as a function of high intensity acoustic drive level. The tetrahedral bubble shape might lead to a smaller mobility than the spherical one [16]. If the acoustic pressure amplitude is increased to 1.3 bars (without inducing explosion), the bubble should be in tetrahedral shape during the part of the acoustic cycle when the pressure is greater than −1.23 bars. The mobility should increase when the bubble shape becomes spherical when the pressure is less than −1.23 bars.

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