

Electron bubbles in liquid ^4He containing a small number of electrons

Wanchun Wei, Zhuolin Xie, and Humphrey J. Maris*

Department of Physics, Brown University, Providence, Rhode Island 02912, USA

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It has been shown that a bubble in liquid helium containing two electrons is unstable against fission. In this paper, we consider the stability of electron bubbles containing 4, 6, or 12 electrons. We find that a bubble with four electrons is unstable at zero pressure and presumably breaks up into single electron bubbles. Our calculation is not accurate enough to determine whether a bubble with six electrons is stable at zero pressure. We find that in liquid ^4He a bubble with 12 electrons is stable over a pressure range from -0.32 to 0.5 bar.

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I. INTRODUCTION

Multielectron bubbles (MEB) in liquid helium were first observed in an experiment by Volodin *et al.* [1]. An electric field was applied to hold a layer of electrons just above the free surface of a bath of liquid helium. The electrons were prevented from entering into the liquid by the repulsion between an electron and helium atoms; this gives an energy barrier of ~ 1 eV [2]. However, at a critical value of the applied electric field, the surface of the liquid became unstable, and electrons entered into the liquid through the formation of bubbles. It was estimated that each of these bubbles contained $10^7 - 10^8$ electrons. Since this pioneering work, there have been several other experiments performed to study multielectron bubbles [3–8], but mostly these experiments have been limited to qualitative observations. The MEB are of interest because they could possibly provide a way to study a number of properties of an electron gas on a curved surface [9].

For an electron bubble containing a large number Z of electrons, it should be a good approximation to treat the electrons classically [classical approximation (CA)]. Then, the energy E of a spherical MEB of radius R in liquid at pressure P is the sum of the energy associated with the Coulomb repulsion of the electrons, the surface energy of the bubble, and the work done against the pressure, i.e.,

$$E = \frac{Z^2 e^2}{2R\epsilon} + 4\pi R^2 \sigma + \frac{4\pi}{3} R^3 P, \quad (1)$$

where σ is the surface tension and ϵ is the dielectric constant (1.0573 at low temperature). This gives an equilibrium radius at zero pressure of [10]

$$R_0 = \left(\frac{e^2 Z^2}{16\pi \sigma \epsilon} \right)^{1/3}. \quad (2)$$

For $N = 10^6$, the radius is 23μ ; this is based on the surface tension of $0.375 \text{ erg cm}^{-2}$ [11].

There are some very interesting questions concerning the stability of these bubbles.

(1) It is clear that if we accept Eqs. (1) and (2) and consider large Z , the bubbles are only metastable, because the total energy is proportional to $Z^{4/3}$, whereas the energy of Z single electron bubbles is proportional to Z .

(2) One can, next, consider the stability of the bubble for small displacements from a spherical shape. Any small displacement $\delta R(\theta, \phi)$ of the sample surface can be expressed as the sum of normal modes, with displacements varying with angle as for the spherical harmonics $Y_{lm}(\theta, \phi)$, i.e.,

$$\delta R(\theta, \phi) = \sum_{lm} \eta_{lm} Y_{lm}(\theta, \phi). \quad (3)$$

Then, to lowest order in the surface displacement, the change in the energy can be written as

$$\delta E = \frac{1}{2} \sum_{lm} \alpha_l |\eta_{lm}|^2. \quad (4)$$

The bubble will be unstable if any of the $\{\alpha_l\}$ are negative. It turns out that if the electrons are treated classically as above, when the pressure is zero, all of the $\{\alpha_l\}$ are positive except for α_1 and α_2 , which are both zero [12,13]. The fact that α_1 is zero is not surprising since any surface displacement with $l = 1$ just corresponds to a translation of the bubble without any change in shape and, therefore, does not change the energy. The result that $\alpha_2 = 0$ raises delicate questions. Guo *et al.* [14] investigated the pressure dependence of α_2 and found that for small P , it varied linearly with P and was negative for $P > 0$. Thus, the bubble was unstable at positive pressures.

(3) The previous discussion has been restricted to the stability of a bubble at rest and with a shape only slightly perturbed from spherical symmetry. Guo *et al.* [14] showed that a moving bubble could be stable at small positive pressures, and Salomaa and Williams [12,13] showed that the presence of a vibration with symmetry $l = 0$ could make a bubble stable with respect to small perturbations with $l = 2$ character.

(4) As a consequence of these results, whether a MEB, which is at rest in liquid at zero pressure, is stable or unstable is presumably determined by corrections to the CA. To discuss this, we first note that although we have been referring to Eqs. (1) and (2) as based on a CA, this cannot be strictly correct. The electric field at the surface gives a finite outward force acting on each electron. If each electron were treated as a classical object of zero size, this force would result in a distortion of the bubble surface allowing the electron to enter the helium. This statement is true, provided we describe the energy of the liquid surface by a surface energy per unit area. Thus, in the CA of Eqs. (1) and (2), it is implicitly assumed that each of the electrons is spread out over some area of the surface so that the electron density per unit area can be taken

*humphrey_maris@brown.edu

as a constant and that the outward electrostatic force on an area can be balanced by the surface tension. Equations (1) and (2) are also based on the approximation that the electrons are confined to a layer of infinitesimal thickness just inside the wall of the bubble. Quantum zero-point motion results in a finite thickness for this layer as considered by Shikin [15] and by Salomaa and Williams [13,16].

(5) As far as we know, there has been very little study of the stability of bubbles containing small numbers of electrons. The stability of a bubble containing two electrons was first investigated by Dexter and Fowler [17]. They performed a variational calculation of the energy of a spherical bubble containing two electrons and showed that this energy was greater than the energy of two single electron bubbles, indicating that a $Z = 2$ bubble is metastable. However, their calculation did not include an investigation of whether there was a way for the shape of the two-electron bubble to evolve so as to lower the energy continuously and break into two. A later investigation showed that this was, in fact, possible [18]. Salomaa and Williams [16] have presented an argument that there is an energy barrier preventing MEB with Z greater than about 20, emitting single electron bubbles.

Multielectron bubbles have been produced by making an electron gas above the free surface of the liquid unstable [1,3,4] by using a cell with a special geometry [5,7] and by an ultrasonic technique [6]. As so far developed, these techniques are not capable of producing bubbles with an accurately controlled number of electrons; this remark applies whether the number Z of electrons in the bubble is small or large. Thus, at the moment, there is no known way to produce a bubble with 12 electrons, for example. However, in a recent experiment by Joseph *et al.* [8] observations of MEB were made with a high-speed camera running at 1000 to 10 000 frames per second. At 2 K and with no pressure applied to the liquid, they recorded the collapse of a 300- μ radius bubble. After a time interval of approximately 7 ms, no remains of the bubble could be seen. This could indicate that all of the electrons had formed single electron bubbles since these would be much too small to be detected. However, it is also possible that bubbles could be produced containing more than one electron, since, if the number of electrons in each bubble were sufficiently small, these would also be too small to be seen. It should be possible to use an ultrasonic method to investigate what is produced in the collapse. It is well-known that bubbles containing a single electron explode at a critical negative pressure of around -2 bars [19,20]. If there are indeed stable bubbles containing several electrons, these should explode at a smaller negative pressure, so it might be possible to confirm their existence by means of an ultrasonic cavitation experiment. In this paper, we investigate the range of pressure over which a bubble containing a small number of electrons is stable.

II. CALCULATION METHOD

To investigate the stability, we need to find an approximation scheme to calculate the total energy of a bubble of a given shape. For the case of two electrons considered in a previous paper [18], it was possible to use a simple diffusion algorithm to solve Schrodinger's equation to find the ground

state of the two interacting particles inside a bubble of arbitrary shape. This required the solution of the diffusion equation in six dimensions. To use this approach for even just four electrons would require a calculation in 12 dimensions, which is impractical.

Consider, first, the minimum Coulomb energy of electrons confined inside a sphere according to classical mechanics. This is a surprisingly complex problem, and there is extensive literature [21–23]. For $Z = 3$, the electrons lie in a plane passing through the center of the sphere, for $Z = 4$, they are at the vertices of a tetrahedron, and for $Z = 6$, they are positioned at equal distances from the origin along the Cartesian axes. For $Z = 12$, the electrons are at the vertices of an icosahedron. For other values of Z , the arrangement of the electrons may have a lower symmetry. For $Z = 8$, for example, it can be shown that the energy is minimal when the electrons are arranged into two groups of four. Each group lies on the corners of a square; one square in the plane $z = 0.5604R$ and the other in the plane $z = -0.5604R$. One square is rotated relative to the other by $\pi/4$ around the z axis, and the distance between the nearest-neighbor electrons in one square is $1.1713R$. The calculation of the quantum correction to the classical result is much easier when the symmetry is high, so we have made calculations for $Z = 4, 6$, and 12 .

We first describe the calculation for the case of four electrons. Given that four electrons confined inside a sphere take on a configuration with tetrahedral symmetry, we assume that the shape of a bubble containing four electrons will also have this symmetry. Let the distance R from the origin to the surface of the bubble in the direction θ, ϕ be written as

$$R(\theta, \phi) = a_0 + \sum_{lm} a_{lm} Y_{lm}(\theta, \phi), \quad (5)$$

where the sum is restricted to $l \geq 2$ and $|m| \leq l$. In order to have the required symmetry, the coefficients $\{a_{lm}\}$ have to satisfy appropriate conditions. For tetrahedral symmetry, the smallest possible value of l is 3. One linear combination of spherical harmonics giving tetrahedral symmetry is obtained with

$$a_{33} = -a_{3\bar{3}} = -a_{30} \left(\frac{2}{5}\right)^{1/2} \quad a_{32} = a_{3\bar{2}} = a_{31} = a_{3\bar{1}} = 0. \quad (6)$$

Using these relations, we can express the bubble shape in the form

$$R(\theta, \phi) = \frac{R_{\max} + R_{\min}}{2} + \frac{R_{\max} - R_{\min}}{2} \times \left[\frac{5}{2} \cos^3 \theta - \frac{3}{2} \cos \theta + \frac{1}{\sqrt{2}} \sin^3 \theta \cos(3\phi) \right], \quad (7)$$

where R_{\max} and R_{\min} are the maximum and minimum values of R , respectively. The bubble has four lobes where the radius reaches the value R_{\max} . These lobes lie in the directions $\hat{e}_1 = (0, 0, 1)$, $\hat{e}_2 = (\frac{2\sqrt{2}}{3}, 0, -\frac{1}{3})$, $\hat{e}_3 = (-\frac{\sqrt{2}}{3}, \frac{\sqrt{2}}{3}, -\frac{1}{3})$, and $\hat{e}_4 = (-\frac{\sqrt{2}}{3}, -\frac{\sqrt{2}}{3}, -\frac{1}{3})$.

One can also construct shapes out of spherical harmonics with larger l , as discussed, for example, by Dudek *et al.* [24]. They find that the next values of l for which tetrahedral symmetry can be satisfied are $l = 7$ and $l = 9$, but we will

restrict the investigation of stability to the set of bubble shapes described by the parameters R_{\max} and R_{\min} .

The problem, then, becomes how to find the energy of four electrons in the shape specified by Eq. (7). We have used the following iterative approach. The linear dimensions of the bubbles of interest are much larger than the Bohr radius $a_0 = \hbar^2/me^2$. Consequently, each electron will have a wave function that is concentrated in one of the lobes, and the overlap of the wave functions of the different electrons will be very small. Hence, we start by placing three electrons localized at the positions $R_{\max}\hat{e}_2$, $R_{\max}\hat{e}_3$, and $R_{\max}\hat{e}_4$, i.e., at the surface of the bubble in the directions of the three lower lobes. We, then, calculate the electrostatic potential acting on the last electron and find the wave function of this electron. To do this, we work with Schrodinger's equation in imaginary time [25] and solve the diffusion equation directly using a finite difference method. This wave function is concentrated in the lobe extending along the positive z axis. Next, we find the average position of this electron; this lies on the z axis at distance ΔR from the end of the lobe, i.e., at position $(R_{\max} - \Delta R)\hat{e}_1$. We now move the three other electrons to the positions $(R_{\max} - \Delta R)\hat{e}_2$, $(R_{\max} - \Delta R)\hat{e}_3$, and $(R_{\max} - \Delta R)\hat{e}_4$ and then repeat the calculation to find a new value of ΔR . Repetition of this procedure gives results for the wave function ψ and the energy that converge rapidly. As an example, we show in Fig. 1, a contour plot of the wave function of this electron in the $x-z$ plane for a bubble with $R_{\max} = 60 \text{ \AA}$ and $R_{\min} = 40 \text{ \AA}$. One can see that the electron is localized in the region within $\sim 30 \text{ \AA}$ of the surface of the bubble. For this geometry, $\Delta R = 16 \text{ \AA}$.

To determine the total energy of the bubble, we calculate the energy E_1 of the last electron and the expectation value $\langle V_1 \rangle$ of the potential energy for this electron. The kinetic energy of this electron is $\langle K_1 \rangle = E_1 - \langle V_1 \rangle$. The potential energy is the sum of the interaction between this electron and the three other electrons. Since there are in total six bonds between the

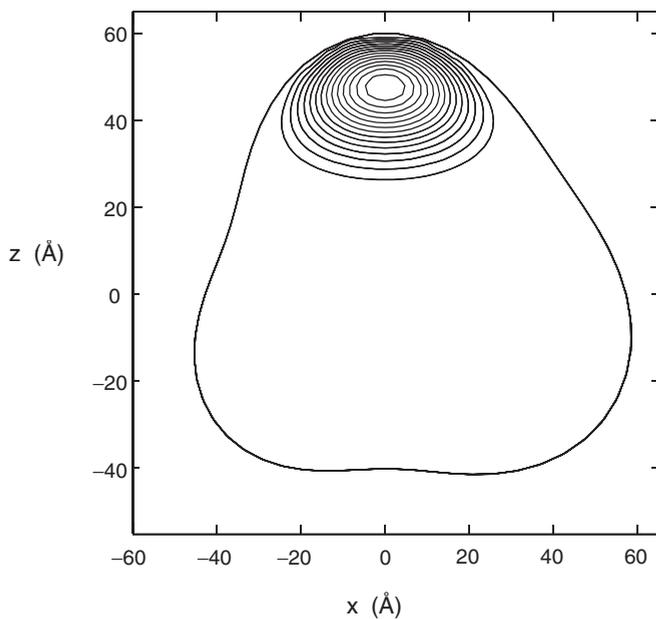


FIG. 1. Contour plot of the wave function in the $x-z$ plane with $y = 0$ for a bubble with four electrons and with $R_{\max} = 60 \text{ \AA}$ and $R_{\min} = 40 \text{ \AA}$.

four electrons, the total energy of the four electrons is then

$$E_{el} = 4\langle K_1 \rangle + 2\langle V_1 \rangle. \quad (8)$$

We calculate E_{el} as a function of the parameters R_{\max} and R_{\min} and then find the total energy of the bubble as

$$E_{\text{total}} = 4\langle K_1 \rangle + 2\langle V_1 \rangle + A\sigma + PV, \quad (9)$$

where A is the surface area of the bubble and V the volume. We then find E_{total} for a range of values of R_{\max} and R_{\min} and look to see if there is a local minimum in the space of these parameters.

For $Z = 6$, the calculation proceeds in a similar way. The lowest value of l needed to create shapes with six lobes and cubic symmetry is four. For $Z = 6$, this gives

$$R(\theta, \phi) = -\frac{R_{\max}}{2} + \frac{3R_{\min}}{2} + 3\frac{R_{\max} - R_{\min}}{2} \times [\sin^4\theta (\cos^4\phi + \sin^4\phi) + \cos^4\theta]. \quad (10)$$

This has lobes extending along the positive and negative Cartesian axes.

For $Z = 12$, we need to have a bubble shape with icosahedral symmetry. The lowest value of l needed to construct a combination of spherical harmonics with icosahedral symmetry is six, and the particular functions needed are Y_{60} , Y_{65} , and Y_{65} . Using these functions, the shape can be expressed in the form

$$R(\theta, \phi) = 0.35714R_{\max} + 0.64286R_{\min} + (R_{\max} - R_{\min}) \times [0.0401785(231\cos^6\theta - 315\cos^4\theta + 105\cos^2\theta - 5) + 1.68750 \cos(5\phi)\sin^5\theta \cos\theta]. \quad (11)$$

The next larger value of l that can be used to construct a shape with icosahedral symmetry is ten.

In performing these calculations, we have not been able to allow for the effect of the polarizability of the liquid, i.e., the results are based on $\epsilon = 1$. Determination of the polarization contribution to the energy would require a calculation of the electric field outside the bubble for each possible position of the electrons inside the bubble as determined by $|\psi|^2$. One can see from Eq. (2) that when $Z \gg 1$, the replacement of ϵ by unity gives a result for the bubble radius that is approximately 2% larger than the correct value.

III. RESULTS

Figures 2–4 show results for the energy of bubbles at zero pressure as a function of R_{\max} and R_{\min} . In each figure, spherical bubbles are indicated by the dashed line. In Fig. 2, we show results for $Z = 4$ and demonstrate that starting from a spherical bubble, the energy can be lowered continuously to reach a geometry in which the minimum radius is very small. With the parameterization that we have chosen in which the bubble shape is completely specified by R_{\max} and R_{\min} , we cannot follow the shape evolution into the state where there are four bubbles each containing a single electron, but it is clear from the contours that the four-electron bubble will evolve into this state. In Fig. 3, we show the results of a calculation of the energy of a bubble with six electrons. There is a shallow

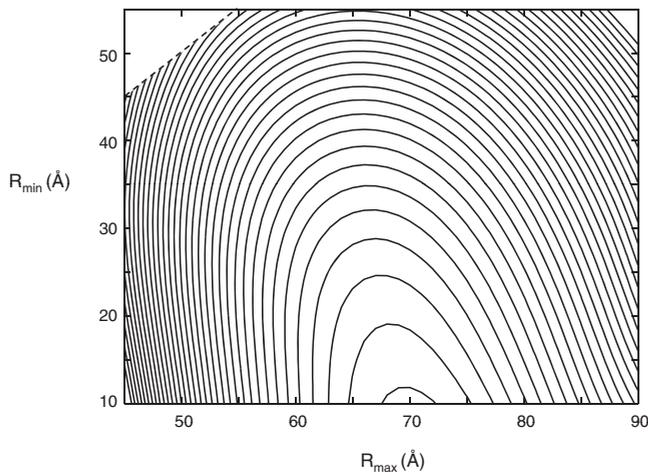


FIG. 2. Contour plot of the energy of a bubble containing four electrons as a function of R_{\max} and R_{\min} . The contour lines are separated by intervals of 2×10^{-14} ergs. The dashed line indicates a spherical bubble.

minimum at $R_{\max} = 91 \text{ \AA}$ and $R_{\min} = 35 \text{ \AA}$. In Fig. 4, we show results for $Z = 12$. A stable configuration is found with $R_{\max} = 130 \text{ \AA}$ and $R_{\min} = 86 \text{ \AA}$.

We, next, investigate the effect of applied pressure. To find the energy, we only need add the term PV to the results already obtained and can then make a new contour plot of the total energy in the $R_{\max} - R_{\min}$ plane. We find that for each of the Z values considered, there is a range of pressure given by $P_{c1} > P > P_{c2}$ in which there is a stable configuration, i.e., a local minimum in the $R_{\max} - R_{\min}$ plane. Results for P_{c1} and P_{c2} are listed in Table I. For pressures positive with respect to P_{c1} , the results suggest that the bubble breaks up into single electron bubbles. For pressures negative with respect to P_{c2} , the bubble expands without limit. In Fig. 5, we show results for R_{\max} and R_{\min} as a function of pressure for the bubble

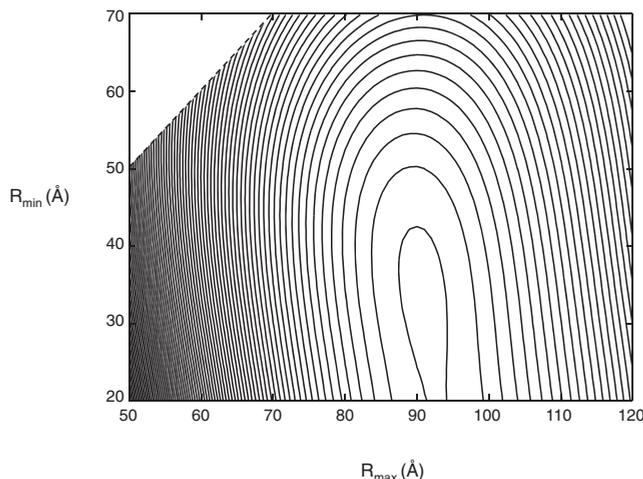


FIG. 3. Contour plot of the energy of a bubble containing six electrons as a function of R_{\max} and R_{\min} . The contour lines are separated by intervals of 3×10^{-14} ergs. The dashed line indicates a spherical bubble.

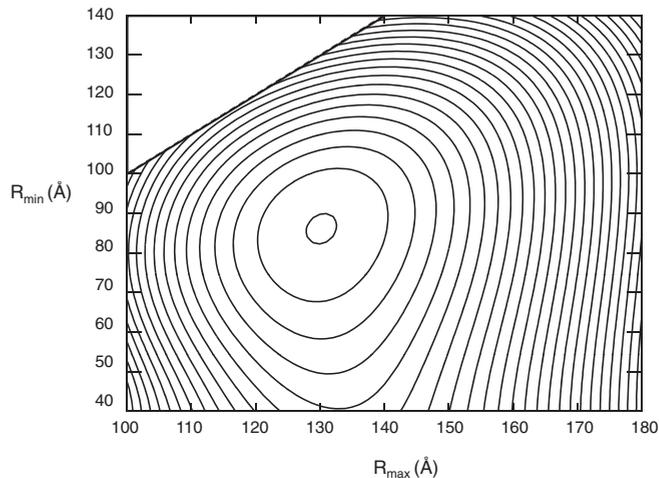


FIG. 4. Contour plot of the energy of a bubble containing 12 electrons as a function of R_{\max} and R_{\min} . The contour lines are separated by intervals of 10^{-13} ergs. The dashed line indicates a spherical bubble.

with 12 electrons. While we can reliably determine P_{c2} to be -0.32 bar, it is much harder to estimate P_{c1} , because when R_{\min} decreases with increasing pressure, we cannot follow the bubble shape to the point where it breaks up. To do this, we would have to add into the expression for the bubble shape a series of spherical harmonics with larger l . The results we list in Table I for P_{c1} are rough estimates of the pressure at which R_{\min} goes to zero, with an uncertainty that may be as large as 0.1 bar. Figure 6 shows the shape of the 12-electron bubble at several pressures.

To improve the accuracy of these calculations, one could use a more general form for the radius as a function of θ and ϕ , including higher-order spherical harmonics but retaining the same overall symmetry. The bubble shape would then be described by three or more parameters, and the calculation would require substantially more computer time. We have made one test of the effect of a larger number of parameters. For $Z = 12$ at zero pressure, we have found that when we use just the spherical harmonics with $l = 6$, there is a stable state with total energy 1.739×10^{-13} ergs. It is straightforward to show that the next higher-order combination of spherical harmonics with icosahedral symmetry is composed of a linear combination of $Y_{10,0}$, $Y_{10,\pm 5}$, and $Y_{10,\pm 10}$ and can be written as

$$f(\theta, \phi) = A_{10} [46189 \cos^{10}\theta - 109395 \cos^8\theta + 90090 \cos^6\theta - 30030 \cos^4\theta + 3465 \cos^2\theta - 63 - 132 \cos(5\phi) \sin^5\theta (323 \cos^5\theta - 170 \cos^3\theta + 15 \cos\theta) + 187 \cos(10\phi) \sin^{10}\theta], \quad (12)$$

TABLE I. Critical pressures for bubbles containing 4, 6, and 12 electrons.

	P_{c1} (bar)	P_{c2} (bar)
$Z = 4$	-0.45	-0.77
$Z = 6$	0.0	-0.60
$Z = 12$	0.5	-0.32

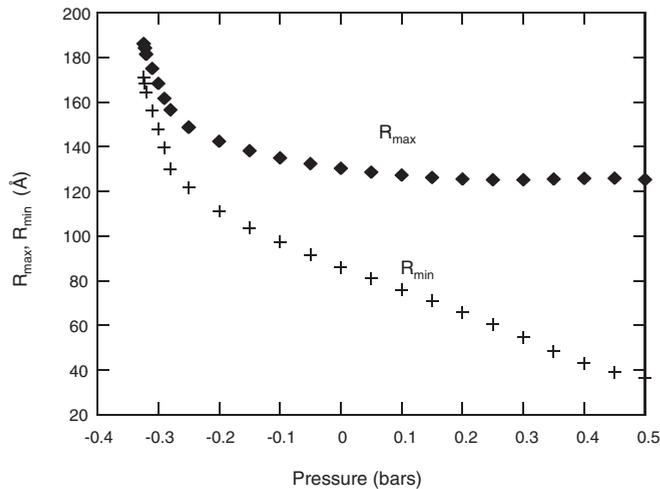


FIG. 5. Variation of the maximum R_{\max} and minimum R_{\min} radius of the 12-electron bubble as a function of applied pressure.

where A_{10} is an arbitrary amplitude. We have investigated the effect of adding this term to the formula for the radius given by Eq. (12) and have calculated the energy as a function of the coefficient A_{10} , while holding the parameters R_{\max} and R_{\min} in Eq. (11) constant. We found that the energy was minimal with $A_{10} = 1.4 \text{ \AA}$ and that by introducing the more general form for the radius, the energy was lowered by only 0.7%. This indicates that the addition of the higher-order harmonics has only a small effect on the stable shape at zero pressure. However, as already mentioned, the effect of higher-order terms will increase rapidly with increasing pressure.

IV. SUMMARY

It would be interesting to extend these calculations to the other quantum liquids, ^3He (lower surface tension) and liquid hydrogen (higher surface tension). There does not appear to be an obvious way to determine the possible stability of bubbles in these liquids without performing a numerical calculation similar to the calculations reported here. The contribution to

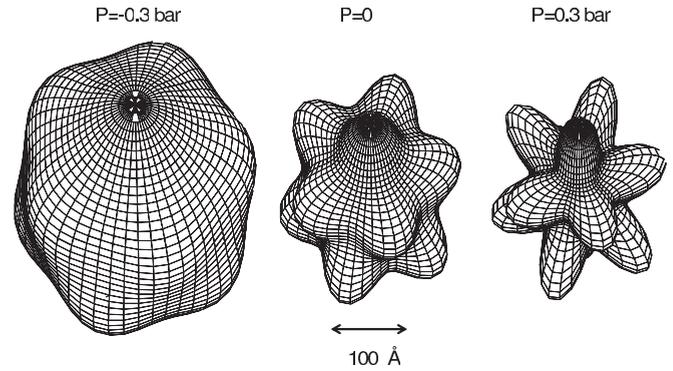


FIG. 6. Calculated equilibrium shape of the 12-electron bubble at pressures of -0.3 , 0 , and 0.3 bars.

the bubble energy from the electrons comes partly from the Coulomb energy and partly from quantum zero-point energy and consequently does not scale with bubble size in a simple way.

The calculations reported here are for $T = 0 \text{ K}$. At finite temperatures, the surface tension will be reduced. In addition, thermal fluctuations can, in principle, enable a bubble to pass over the barrier preventing break up into single electron bubbles. However, one can see from the figures showing bubble energies that at temperatures of a few Kelvin, kT is very small compared to the energy scale of the bubble.

In summary, our results indicate that electron bubbles containing a small number of electrons can be stable in agreement with the earlier ideas of Salomaa and Williams [13]. It may be possible to detect these objects using ultrasonic cavitation. The calculations indicate that there exist bubbles with small value of Z that are stable at zero pressure but can result in cavitation at small negative pressures.

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