Stability of multielectron bubbles in liquid helium

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The stability of multielectron bubbles in liquid helium is investigated theoretically. We find that multielectron bubbles are unstable against fission whenever the pressure is positive. It is shown that for moving bubbles the Bernoulli effect can result in a range of pressures over which the bubbles are stable.

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I. INTRODUCTION

Multielectron bubbles in liquid helium were first observed by Volodin *et al.*[1] In their experiment a layer of electrons was held in place just above the free surface of a bath of liquid helium by an electric field. The field was produced by a positive voltage applied to an electrode immersed in the liquid. The electrons remained outside the helium because for an electron to enter liquid helium it has to overcome a potential barrier of height approximately 1 eV.[2] When the field reached a critical value, the surface of the liquid became unstable and a large number of electrons entered into the liquid through the formation of bubbles. Each of these bubbles typically contained $10^7 \sim 10^8$ electrons. The multielectron bubbles are of interest because they could possibly provide a way to study a number of properties of an electron gas on a curved surface.[3]

As a first approximation, one can consider that the radius of a spherical multielectron bubble (MEB) is such as to minimize the sum of the energy associated with the Coulomb repulsion of the electrons and the surface energy of the bubble. This gives an equilibrium radius of

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

where Z is the number of electrons, σ is the surface tension of helium (0.36 erg cm^{-2} at 1.3 K),[4] ϵ is the dielectric constant (1.0573 at low temperature), and the applied pressure has, for the moment, been taken to be zero. Thus, for example, for $Z=10^7$ the radius is 106 μm.

So far, there have been a very limited number of experimental studies of these bubbles.[1,5–7] In this paper we first consider the stability of an MEB that is at rest in liquid (Sec. II). We find that, at least when the simplest model of the energy of the electron system is used, the bubble is unstable against fission whenever the applied pressure is positive. In Sec. III we investigate how the stability of a bubble is changed when it is moving through the liquid. We have been able to determine the region in the pressure-velocity plane where the bubble is stable. In the conclusion we mention some interesting differences between the behavior of multielectron bubbles and single electron bubbles. The numerical calculations in this paper are all for helium-4.

II. STABILITY OF BUBBLES AT REST

Since MEBs were first observed, there have been several theoretical investigations of the stability of these objects. The first discussion was given by Shikin[8] and further analysis was given by Salomaa and Williams,[9–11] and Tempere and co-workers.[3,12–14] In the simplest model, the electrons are taken to be distributed over the inner surface of the bubble such that the electric field is everywhere exactly normal to the surface. This ensures that the charge distribution is in equilibrium. The electrons are treated classically and so they are localized at the surface in a layer of zero thickness (see below). Thus the total energy of the bubble is taken to be

$$E = E_S + E_V + E_C. \tag{2}$$

Here $E_S = \sigma S$ is the surface energy, where S is the surface area and σ is the surface tension, $E_V = PV$ is the volume energy (where P is the applied pressure and V is the bubble volume), and E_C is the Coulomb energy given by

$$E_C = \int \frac{\epsilon \mathcal{E}^2}{8\pi} dV. \tag{3}$$

Since the electrons can move freely around the surface, the field $\mathcal{E}$ inside the bubble must be zero, and so the integral in Eq. (3) can be restricted to the region outside the bubble. If the bubble is spherical, the bubble radius that gives the minimum value of the energy is the solution of the equation,

$$R_0 = \left(\frac{Z^2 e^2}{16\pi\epsilon\sigma + 8\pi\epsilon P R_0} \right)^{1/3}. \tag{4}$$

For zero applied pressure, this gives the total energy of an MEB as

$$E = \frac{3}{2} \left(\frac{2\pi Z^4 e^4 \sigma}{\epsilon^2} \right)^{1/3}. \tag{5}$$

Since the energy is proportional to $Z^{4/3}$ the energy is always reduced if the bubble breaks into two. Hence, in the discussion of stability given here, we are not considering whether the energy of the bubble can be lowered if it breaks into pieces but we are trying to determine whether there is an energy barrier that prevents the bubble from breaking.

To consider whether the spherical shape is stable, we write

$$R(\theta, \phi) = R_0 \left\{ 1 + \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \eta_{lm} Y_{lm}(\theta, \phi) \right\}, \tag{6}$$

where $\eta_{l,-m} = \eta_{lm}^*$. It is straightforward to show that to second order in the parameters η_{lm}, the three contributions to the energy can be written as

$$E_S = 4\pi\sigma R_0^2 \left\{ 1 + \frac{1}{\sqrt{\pi}}\eta_{00} \right.$$
$$\left. + \frac{1}{8\pi}\sum_{l=0}^{\infty}\sum_{m=-l}^{l}(l^2+l+2)|\eta_{lm}|^2 \right\}, \quad (7)$$

$$E_V = \frac{4\pi}{3}PR_0^3 \left\{ 1 + \frac{3}{2\sqrt{\pi}}\eta_{00} \right.$$
$$\left. + \frac{3}{4\pi}\sum_{l=0}^{\infty}\sum_{m=-l}^{l}|\eta_{lm}|^2 \right\}, \quad (8)$$

$$E_C = \frac{Z^2 e^2}{2\epsilon R_0} \left\{ 1 - \frac{1}{2\sqrt{\pi}}\eta_{00} + \frac{1}{4\pi}\eta_{00}^2 \right.$$
$$\left. - \frac{1}{4\pi}\sum_{l=0}^{\infty}\sum_{m=-l}^{l}l|\eta_{lm}|^2 \right\}. \quad (9)$$

Hence the total energy is

$$E = 12\pi\sigma R_0^2 + \frac{16\pi}{3}P\,R_0^3 + \sum_{l=0}^{\infty}\frac{1}{2}\alpha_l\sum_{m=-l}^{l}|\eta_{lm}|^2, \quad (10)$$

where the spring coefficients α_l are given by

$$\alpha_0 = 6\sigma R_0^2 + 4PR_0^3, \quad (11)$$

and

$$\alpha_l = (l-2)(l-1)\sigma R_0^2 - 2(l-1)PR_0^3, \quad (12)$$

for $l \geq 1$. From this one can see that the bubble is stable against spherically symmetric perturbations provided that $6\sigma R_0^2 + 4PR_0^3 > 0$. This leads to the condition $P > P_c$ where

$$P_c = -\left(\frac{27\pi\epsilon\sigma^4}{2Z^2e^2}\right)^{1/3}. \quad (13)$$

For $l=1$, the spring coefficient α_1 is zero; this is to be expected since a perturbation of the form $\eta_{1m}Y_{1m}(\theta,\phi)$ corresponds to a simple translation of the bubble in some direction. For $l=2$ the spring constant α_2 is zero if the pressure is zero, and so this analysis of the effect of small perturbations to the initial spherical shape does not determine the stability of the bubble. The higher l spring constants are all positive at zero pressure but each becomes negative if the pressure is increased to a sufficiently positive value. It was noted by Tempere *et al.*[12] that if the pressure is negative (but not negative with respect to P_c), all of the spring constants will be positive[15] and so the bubble must be stable.

The stability of the bubble at zero pressure is of special importance, since in the experiments that have been performed so far, there has been no applied pressure, apart from the very small hydrostatic pressure due to the distance between the bubble and the free surface. At zero pressure α_2 is zero, and so we need to go beyond the lowest order in perturbation theory in order to investigate the stability of an MEB at zero pressure. One approach would be to calculate the terms in the energy that are fourth order in the η_{lm} parameters. Instead we have performed numerical calculations of the total energy as a function of bubble shape.

To do this, we describe the shape of the bubble using Eq. (6), but now we do not restrict the parameters η_{lm} to being small. When the bubble shape changes, the electrons will redistribute themselves over the surface so as to minimize the energy and to make the electric field inside the bubble zero. For each choice of shape we use the finite element method[16] to compute the surface charge distribution and the Coulomb energy. The simulation uses 1280 triangle patches. We start with a spherical shape and we vary the parameters η_{lm} to see if a state of lower energy can be reached without passing over a barrier. We have done this using a maximum value of five for l in Eq. (6). This process was then repeated for a series of different pressures. We also performed similar calculations with a maximum value of $l=15$ but taking only $m=0$. Both procedures gave the same results for the stability.

The result of this investigation is that for all positive pressures there is no barrier to fission, whereas for negative pressures there is a barrier. This result holds for all values of Z. To illustrate the path to fission, we describe the results obtained for a simplified calculation, in which only $l=0$ and $l=2$ contributions are retained. Thus we write

$$R(\theta,\phi) = a_0 + a_2(3\cos^2\theta - 1). \quad (14)$$

Within this simplified model, fission occurs when $a_2 = a_0$ and the bubble develops a hole along the z axis when $a_2 = -a_0/2$, i.e., takes on a donut shape. In Fig. 1(a), we show examples of contour plots of the energy in the a_0-a_2 plane. The pressure is -0.03 mbar and $Z=10^6$. There is a stable minimum with a_2 equal to zero, i.e., the bubble is spherical. When the pressure is zero [Fig. 1(b)], there is still a point in the plane at which the energy of the bubble is stationary with respect to both a_0 and a_2 (at $a_0 = 23.8$ μm and $a_2 = 0$), but it is now possible to reach the fission line from this point without passing over any energy barrier. Note that along this path there is, of course, an increase in the value of a_2 but also a substantial decrease in a_0. Once the pressure becomes positive [see, for example, Fig. 1(c)], there is no point in the a_0-a_2 plane where the energy is stationary.

These results can be compared with the earlier calculations by Tempere *et al.*[14] who also investigated the stability against fission. They used an ingenious method, in which the bubble was described by six parameters chosen so that the shape of a bubble undergoing fission could consist of two spheroids connected by a hyperboloidal neck. The choice of parameters was such that the bubble could vary from a single sphere, to an ellipsoid, and then all the way to separated spheres. They minimized the total energy of the bubble by adjusting these parameters subject only to the constraint that the total length L of the bubble had to have a given value. They then investigated how the total energy varied with L starting from a value of L equal to $2R_0$. If the energy decreased monotonically as L increased from $2R_0$ to a large value, this indicates that the MEB is unstable against fission. If the energy first increases before decreasing, this indicates that the bubble is stable. To simplify the calculation, Tempere and co-workers made the approximation that the charge density was uniform over the surface of the bubble. They concluded that at zero pressure even though there is a mode of deformation (the $l=2$ mode), which can grow without in-
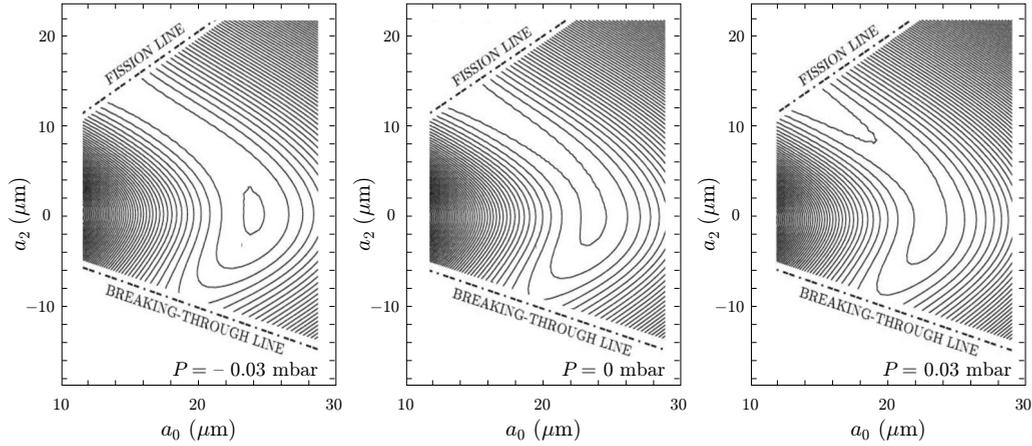
FIG. 1. Contour lines of constant energy for a multielectron bubble containing 10^6 electrons for three different pressures. The energy spacing between contour lines is 0.05 eV. The energy is shown as a function of the parameters a_0 and a_2 as defined in Eq. (14). The electrons are distributed over the bubble surface so as to minimize their energy.

creasing the energy of the bubble, there should be an energy barrier that prevents fission,[17] whereas we find no barrier.

This difference in the results arises from the treatment of the charge distribution on the bubble. If the bubble is assumed to have surface charge density that remains uniform when the shape changes, it is straightforward to show that the Coulomb energy for small changes from the equilibrium spherical shape is

$$E_C = \frac{Z^2 e^2}{2\epsilon R_0} \left\{ 1 - \frac{1}{2\sqrt{\pi}} \eta_{00} \right. \\ \left. - \frac{1}{4\pi} \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \frac{l^2 + 3l - 1}{2l+1} |\eta_{lm}|^2 \right\}. \quad (15)$$

In this case the spring constant α'_l for the lth mode (considering only $l \geq 1$) becomes

$$\alpha'_l = (l^2 + l + 2)\sigma R_0^2 + 2P R_0^3 - \frac{l^2 + 3l - 1}{2l+1} \frac{Z^2 e^2}{4\pi\epsilon R_0}. \quad (16)$$

Comparing this with the spring constant α_l when the charge redistributes, Eq. (12) gives

$$\alpha'_l = \alpha_l + \frac{(l-1)^2}{2l+1} \frac{Z^2 e^2}{4\pi\epsilon R_0}. \quad (17)$$

Thus for all modes, except $l=0$ and $l=1$, making the approximation of a uniform surface charge gives an increase in stiffness and makes it harder for the bubble to undergo fission. The increase in stiffness is to be expected since a redistribution of surface charge can only lower the total energy. In Fig. 2, we show energy contour lines in the a_0–a_2 plane for an MEB with 10^6 electrons at zero pressure calculated by taking a uniform surface charge. One can see that within this approximation the spherical bubble is stable.

There are several physical effects that are not included in the simplified model used so far. It is possible that allowance for these effects would change the stability of an MEB at zero pressure. A more detailed consideration of the Coulomb energy (the total electron energy, to be more precise) for a

spherical bubble was given by Salomaa and Williams[9,10] using the density-functional formalism of Hohenberg and Kohn.[18] This makes it possible to include the kinetic, exchange, and correlation energies. However, how these extra contributions affect the spring constants is not clear and is difficult to calculate. Salomaa and Williams showed that these extra contributions to the energy make a very small contribution to the energy when Z is large. For example, for $Z = 10^8$ the extra terms make a contribution that is roughly 4000 times smaller than the form $Z^2 e^2 / 2\epsilon R_0$ for the energy used in the simple model. The calculation could also be improved, for example, by using a density-functional theory to treat the surface of the liquid helium and by allowing for the
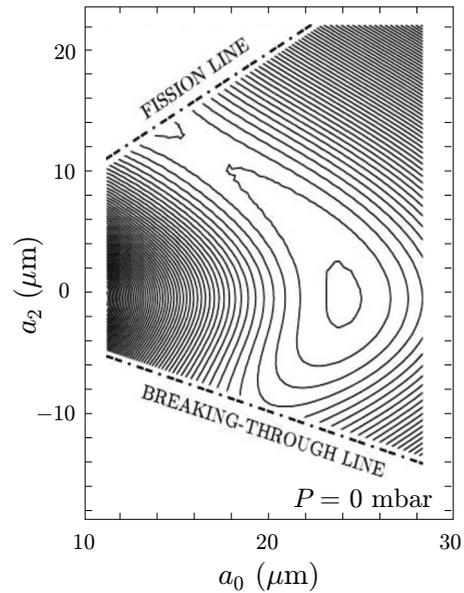

FIG. 2. Contour lines of constant energy for a multielectron bubble containing 10^6 electrons at zero pressure. The energy spacing between contour lines is 0.05 eV. The energy is shown as a function of the parameters a_0 and a_2 as defined in Eq. (14). The electrons are uniformly distributed over the surface area of the bubble.

penetration of the electron wave function into the liquid. All of these effects appear to be a very small correction to the total energy and hence are unlikely to change the spring constants by a large amount. However, it is important to note that even a small correction could lead to a positive value for α_2, which would in turn lead to a finite (but small) energy barrier against fission. As an example, consider the corrections that arise as a result of using a density-functional scheme to describe the helium. For a bubble with radius large compared to the thickness of the liquid-vapor interface, the first correction to the energy can be represented by considering the surface tension σ to contain a correction that is proportional to κ, the sum of the principle curvatures of the surface. Based on a simple density-functional scheme used previously,[19] it is straightforward to show that the correction to the surface tension is $\Delta\sigma = \sigma'\kappa$, where

$$\sigma' = 0.9 \times 10^{-8} \text{ erg cm}^{-1}, \tag{18}$$

and the sign of the correction is such that the surface tension is increased for a concave surface of the liquid. Inclusion of this term changes the total energy by an amount ΔE, which for a bubble at zero pressure is given by

$$\frac{\Delta E}{E} = \frac{0.08}{Z^{2/3}}. \tag{19}$$

It is straightforward to show that the spring constant for an $l=2$ deformation at zero pressure now becomes

$$\alpha_2 = 12\sigma' R_0. \tag{20}$$

Because α_2 is now positive at zero pressure, there will be a barrier against fission. However clearly for large Z (e.g., $Z \sim 10^8$), this barrier will be very small.

Another effect that may influence the stability was pointed out by Williams and Salomaa.[9–11] They show that if the bubble is undergoing a spherically symmetric oscillation of finite amplitude (an oscillation involving η_{00}) then there is an increase in the effective spring constant for oscillations with $l=2$, i.e., the spherically symmetric oscillations tend to stabilize the bubble. It would be interesting to extend the present calculations to include such dynamic effects. For example, one could start with a bubble in a nonequilibrium configuration corresponding to some initial value of the $\{\eta_{lm}\}$ coefficients and their time derivatives $\{\dot{\eta}_{lm}\}$, and then one could calculate how the bubble shape evolves as time elapses. However, this is a significantly more complex computation and would require both a large number of calculations of the electron distribution inside the bubble and also a calculation of the motion of the liquid surrounding the bubble.

III. STABILITY OF MOVING BUBBLES

The above results indicate that one way to stabilize an MEB is to produce it in liquid that is under a small negative pressure. We now consider an alternate way to maintain a stable bubble. A bubble moving through the liquid will be affected by the local pressure change associated with the liquid moving around it. For a spherical bubble moving at velocity v through an incompressible inviscid fluid with density ρ, the Bernoulli effect results in a pressure variation over the surface of the bubble, which is given by[20]

$$P(\theta) = P_0 + \frac{1}{8}\rho v^2(9 \cos^2 \theta - 5)$$
$$= P_0 - \rho v^2 \sqrt{\frac{\pi}{4}} Y_{00}(\theta, \phi) + \rho v^2 \sqrt{\frac{9\pi}{20}} Y_{20}(\theta, \phi). \tag{21}$$

For a bubble in liquid that is at zero pressure far removed from the bubble ($P_0 = 0$), this changes the shape of the bubble in two ways. The term proportional to Y_{00} by itself would provide a negative pressure around the surface of the bubble and since bubbles are stable at negative pressure, this contribution serves to stabilize the bubble. The second term gives a positive pressure at the poles of the bubble and a negative pressure around the waist. This pressure distribution will distort a spherical bubble so as to make the parameter η_{20} in Eq. (6) [or a_2 in Eq. (14)] to be negative. This tends to stabilize the bubble since, as can be seen from Fig. 1, for fission to occur a_2 has to become positive.

We have performed computer simulations in order to find the shape of moving bubbles and the range of velocity and pressure for which they are stable. We start with a guess at the bubble shape and then calculate the charge distribution on the surface. This then gives the pressure $\Delta P_{el}(\theta)$ exerted on the surface by the electrons. We then find the flow in the liquid. To do this, we expand the velocity potential as

$$\Phi(\theta) = \sum_l B_l P_l(\cos \theta) r^{-l-1}, \tag{22}$$

where B_l are some coefficients and the sum includes terms up to $l=20$. The coefficients are determined so as to give a velocity distribution in the liquid such that in the frame of reference of the moving bubble, the liquid velocity at the bubble surface in the direction normal to bubble surface is as close to zero as possible. This gives a pressure at the bubble surface of

$$P_0 + \Delta P_B(\theta), \tag{23}$$

where P_0 is the pressure in the bulk liquid far removed from the bubble and $\Delta P_B(\theta)$ is the Bernoulli pressure. The net inward force acting on unit area of the bubble surface is then

$$P_0 + \Delta P_B(\theta) + \sigma\kappa - \Delta P_{el}(\theta), \tag{24}$$

where κ is the sum of the principle curvatures of the surface and $\Delta P_{el}(\theta)$ is the outward pressure exerted by the electrons. Each part of the bubble surface is then moved inward a distance proportional to this force, and the process repeats until the equilibrium shape is found. This calculated shape is only stable against axial symmetric variations. In order to test its overall stability including nonaxial symmetric variations, we added some nonaxial symmetric perturbations on the shape and repeated the simulation mentioned above. However this time, the fluid potential is expanded by spherical harmonics with maximum $l=5$ and all $m \neq 0$. For an MEB with $Z = 10^6$, the stable shapes for three velocities at zero P_0 are shown in Fig. 3. We are able to perform the numerical cal-
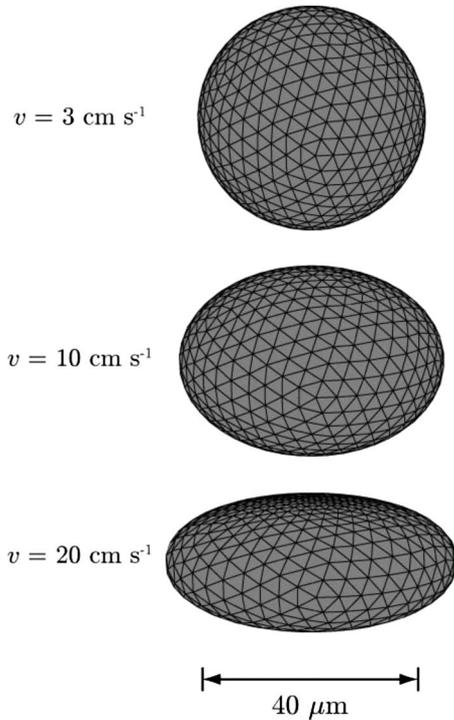
FIG. 3. The shape of a multielectron bubble containing 10^6 electrons for bubble velocities of 3, 10, and 20 cm s^{-1}. The pressure at large distance from the bubble is zero.

FIG. 4. Distance R_{pole} from the bubble center to the poles and the radius R_{waist} of the waist as a function of the bubble velocity. These results are for a bubble containing 10^6 electrons moving through liquid in which the pressure at large distance from the bubble is zero.

culation until the bubble becomes concave at the poles. This is shown by the dashed line in Fig. 5.

In Fig. 4, the distance R_{pole} from the bubble center to the pole and the radius R_{waist} of the waist are shown as a function of the velocity. In Fig. 5 we show a plot of the region in the pressure-velocity plane in which the bubble is stable. This region is bounded by two lines. For small velocities there is a critical positive pressure at which the bubble undergoes fission. At negative pressures the bubble becomes unstable against expansion. For zero velocity this expansion is isotropic.

Note that the change in the shape of the bubble even for a small velocity is surprisingly large. This comes about simply because the Bernoulli pressure contains a finite term varying with angle $Y_{20}(\theta, \phi)$ but the spring constant α_2 for this pressure component is zero. Thus, for an MEB the changes in R_{pole} and R_{waist} are linearly proportional to the bubble velocity, whereas for a gas bubble in liquid the spring constant α_2 is finite and so the changes in dimensions are proportional to the square of the velocity.

The region of stability of bubbles containing a different number of electrons can be found by scaling the results shown in Fig. 5. The instability pressure $P_{instab}(Z, v)$ can be written in the form

$$P_{instab}(Z, v) = AZ^{-2/3} f(Bv^2 Z^{2/3}), \qquad (25)$$

where f is a dimensionless function, $A = (\sigma^4 \epsilon / e^2)^{1/3}$, and $B = \rho(e^2 / \sigma^4 \epsilon)^{1/3}$. Thus, for zero velocity the critical negative pressure, at which a bubble becomes unstable, is proportional to $Z^{-2/3}$, and at zero applied pressure the critical ve-
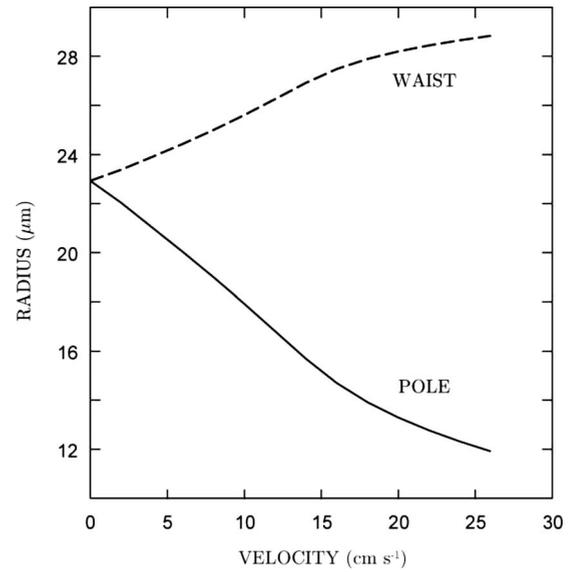
locity, at which the bubble becomes concave at the poles occurs, is proportional to $Z^{-1/3}$. This scaling law can also be used to give the instability pressure for an electron bubble in helium-3 since the only parameter of the liquid that enters is the surface tension.

We note that in this paper we have treated the liquid as inviscid although, of course, helium above the lambda point has a finite viscosity and below the lambda point the liquid still has a normal fluid component. At sufficiently low temperatures the density of the normal fluid becomes very small and, in addition, the mean-free path of the excitations making up the normal fluid becomes comparable to the radius of an MEB. Under these conditions, it appears that the only
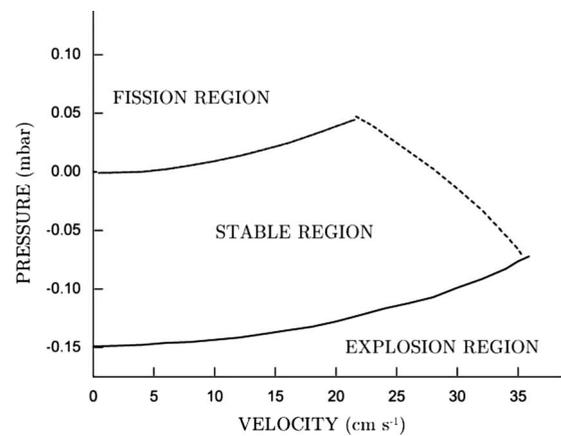

FIG. 5. Plot of the region in the pressure-velocity plane in which a MEB containing 10^6 electrons is stable. The region is bounded by the lines on which the two different types of instability occur as described in the text. Along the dashed line, the bubble becomes concave at the poles and the numerical calculations become inaccurate.

effect of the normal fluid is to determine the mobility of an MEB. There should be no effect on the shape change or the stability. For a bubble with $Z=10^6$ the mean-free path of the thermal excitations becomes equal to the radius at around 0.6 K and at this temperature the normal fluid density is less than the total density by a factor of 4×10^{-5}. However as far as we are aware, there have been no experiments with MEBs at low temperature.

At high temperatures where the helium is in the normal state, the situation is not so clear. It is known that when the Reynolds number is large (but not so large that the flow becomes turbulent) the viscosity results in a thin boundary layer on the surface of the bubble and the pressure at the bubble surface is close to the value that would result from potential flow.[21] This general idea would suggest that the inviscid approximation should give reliable results for the stability of MEBs over a wide range of the Reynolds number. To determine this range, one could calculate the effect of viscosity to the shape and drag on the gas bubbles moving through the liquid using the method developed by Li and Yan.[22] We have not attempted to do this. We note that, Albrecht and Leiderer[7] in their experiments at 3.5 K found that the mobility of the MEBs was between one and two orders of magnitude smaller than what is expected on the basis of ordinary hydrodynamics. The reason for this is unknown. In the experiments of Volodin *et al.*[1] and Khaikin,[5] which were performed at 1.3 K, velocities of the order of 10^4 cm s^{-1} were reported. At these velocities the normal fluid component would be in the turbulent regime and the bubble is moving so fast that it should lose energy through the production of quantized vortex rings.

IV. CONCLUSION

We have examined the stability of multielectron bubbles in liquid helium and found that stationary bubbles at positive pressures are unstable. We show that because of the Bernoulli effect moving bubbles can be stable even at small positive pressures.

It is interesting to compare the results obtained here for an MEB with the behavior of a single electron bubble (SEB). For an SEB it is essential to treat the electron using quantum mechanics. The shape is determined by a balance between surface tension, the quantum pressure exerted by the electron on the inside of the bubble wall, and the Bernoulli pressure acting on the outside. The change in the shape of an SEB was calculated by Guo and Maris.[23] They found that for an SEB bubble both R_{waist} and R_{pole} increase with increasing velocity and the difference between them is small so the bubble remains approximately spherical. This is in contrast to the results for the MEB where R_{waist} increases and R_{pole} decreases as shown in Fig. 4. The different behavior is related to the different "elastic" behavior of the bubble contents. For an SEB there is a large energy increase associated with a change in shape of the electron wave function even if there is no change in the volume. This is in contrast to the behavior of the electron energy (i.e., the Coulomb energy) for the MEB. From Eq. (9), one sees that the Coulomb energy actually *decreases* as the square of η_{20}. Thus, the response of an SEB to a surface pressure varying as Y_{2m} is much smaller than the response of an MEB and it is for this reason that the SEB bubble remains nearly spherical as the velocity increases, whereas the MEB undergoes a large distortion.

ACKNOWLEDGMENT

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