Theory of the stability of multielectron bubbles in liquid helium

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Abstract.
We have performed numerical calculations to investigate the stability of multi-electron bubbles in liquid helium-4. In our calculations, for each bubble shape considered we find the equilibrium distribution of the electron gas on the inside surface of the bubble. For a stationary bubble, we find that whenever the pressure in the liquid is positive the bubble is unstable against fission into two bubbles. We find that a moving bubble can be stable within a certain region of the pressure-velocity plane. At the high pressure boundary of this region, the bubble undergoes fission, and on the low pressure boundary the bubble explodes.

Electrons above the free surface of a bath of liquid helium will remain outside the helium because of a 1 eV potential barrier [1]. If a positive voltage is applied to an electrode immersed inside the liquid, when the field reaches a critical value, the surface of the liquid becomes unstable and a large number of electrons enter into the liquid through the formation of multi-electron bubbles (MEB). Each of these bubbles typically contain $10^7 \sim 10^8$ electrons with radii about 100 μm [2].

There have been several theoretical investigations of the stability of MEB [3, 4]. In the simplest model, the electrons are treated as classical particles with Coulomb repulsion and so are localized at the inner surface of bubble in a layer of zero thickness. The distribution of electrons is such that the electric field is everywhere exactly normal to the surface. This ensures that the charge distribution is in equilibrium. The stable bubble shape is attained by minimizing the total energy

$$E = E_S + E_V + E_C. \tag{1}$$

Here $E_S = \sigma S$ is the surface energy with S the surface area and σ the surface tension (0.36 erg cm^{-2} at 1.3 K), $E_V = PV$ the volume energy with P the applied pressure and V the bubble volume. E_C is the Coulomb energy given by $E_C = \int(\epsilon\mathcal{E}^2/8\pi)\mathrm{d}V$, where ϵ is the dielectric constant (1.0573 at low temperature), and the electric field $\mathcal{E}$ is nonzero only outside the bubble. For a spherical bubble of Z electrons, the radius that gives the minimum value of the energy is the solution of the equation $R_0^3(8\pi\epsilon P R_0 + 16\pi\epsilon\sigma) = Z^2e^2$.

To consider whether the spherical shape is stable, we introduce the shape parameters η_{lm} and write

$$R(\theta,\phi) = R_0\left\{1 + \sum_{l=0}^{\infty}\sum_{m=-l}^{l}\eta_{lm}Y_{lm}(\theta,\phi)\right\}. \qquad (\eta_{l,-m} = \eta_{lm}^*) \tag{2}$$

If the η_{lm} are small, one can show that to the second order, the total energy is

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

where the spring coefficients κ_l are given by

$$\kappa_l = \begin{cases} 6\sigma R_0^2 + 4PR_0^3, & (l = 0) \\ (l-2)(l-1)\sigma R_0^2 - 2(l-1)PR_0^3. & (l \geq 1) \end{cases} \tag{4}$$

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 $P > P_c = -\left(27\pi\epsilon\sigma^4/2Z^2e^2\right)^{1/3}$. For $l = 1$, the spring coefficient κ_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 κ_2 is zero if the pressure is zero, and negative if the pressure is (arbitrarily small) positive. This means that a spherical bubble within this pressure range is unstable against an $l = 2$ shape change. 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. In addition, if a negative pressure is applied (but not negative with respect to P_c), all of the spring constants will be positive (excluding κ_1 which is always zero) and so the bubble should be stable [4].

The stability of the bubble at zero or small positive pressure is of especial 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 the bubble is below the free surface. Although the above perturbation analysis shows the instability of a spherical bubble against the $l = 2$ mode, it does not rule out the possible existence of a non-spherical stable shape (energy local minimum) at some finite η_{lm}. So we have performed numerical calculations by the finite element method [5] without restricting the parameters η_{lm} to being small. For each shape we allow the electrons to redistribute themselves so as to minimize the energy and to make the electric field inside the bubble zero.

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 results obtained for a simplified calculation in which only $l = 0, 2$ and $m = 0$ contributions are retained. Thus we write

$$R(\theta, \phi) = a_0 + a_2(3\cos^2\theta - 1). \tag{5}$$

Within this simplified model, fission occurs when $a_2 = a_0$; and the bubble develops a hole along the z-axis, i.e., takes on a donut shape, when $a_2 = -a_0/2$. In Fig. 1 we show examples of contour plots of the energy in the a_0-a_2 plane for $Z = 10^6$. For a pressure of -0.03 mbar (Fig. 1(a)), 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 (at 23.8 μm) and a_2 (at 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 (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.* [4], who also investigated the stability against fission. To simplify the calculation, they made the approximation that the charge density was uniform over the surface of the bubble and concluded there exists an energy barrier which prevents fission, whereas we find no barrier. To understand

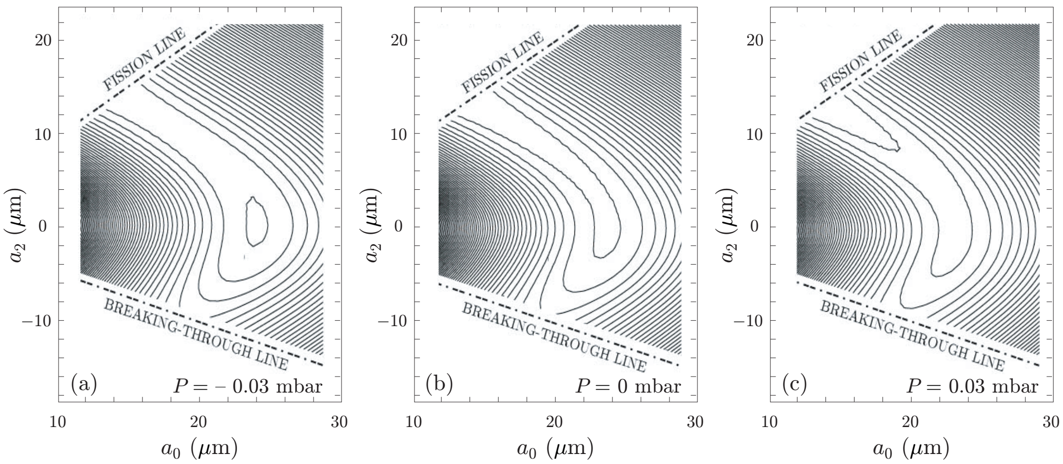

Figure 1. Contour lines of constant energy for an MEB containing 10^6 electrons for three different pressures. The energy spacing between contour lines is 0.05 eV.

this difference, we redid the perturbation analysis by assuming that when the shape changes the surface charge distribution remains uniform. This gave new spring constants κ_l'

$$\kappa_l' = \kappa_l + \frac{(l-1)^2}{2l+1}\frac{Z^2e^2}{4\pi\epsilon R_0}, \qquad (l \geq 1) \tag{6}$$

which are larger than the spring constants κ_l found when it is assumed that the charge redistributes (except for the $l = 1$ mode which remains zero). This increase in stiffness is to be expected since a redistribution of surface charge can only lower the energy.

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 a 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 [6]

$$P(\theta) = P_0 + \frac{1}{8}\rho v^2(9\cos^2\theta - 5) = P_0 - \rho v^2\sqrt{\frac{\pi}{4}}Y_{00}(\theta,\phi) + \rho v^2\sqrt{\frac{9\pi}{20}}Y_{20}(\theta,\phi). \tag{7}$$

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 uniform 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. (2), or a_2 in Eq. (5), 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.

In order to find the shape of moving bubbles and the range of velocity and pressure for which they are stable, we have performed computer simulations by balancing the surface forces due to the potential flow and the electron repulsion. For an MEB with $Z = 10^6$ the stable shapes for three velocities at zero P_0 are shown in Fig. 2. In Fig. 3 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.

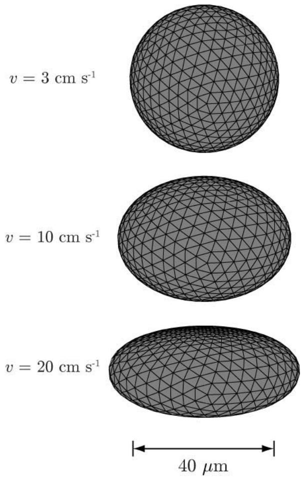

Figure 2. The shape of an MEB containing 10^6 electrons for three different velocities. The pressure far away from the bubble is zero.

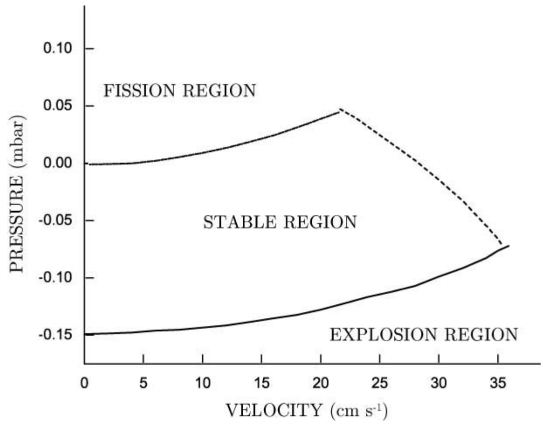

Figure 3. Plot of the stable region in the pressure-velocity plane for an MEB containing 10^6 electrons. It is bounded by the lines on which the two different types of instability occur. Along the dashed line the bubble becomes concave at the poles and the numerical calculations become inaccurate.

At negative pressures the bubble becomes unstable against expansion. For zero velocity this expansion is isotropic. We are only able to perform the numerical calculation until the bubble becomes concave at the poles (shown by the dashed line).

We note that in the above calculation 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 effect of the normal fluid is to determine the mobility of an MEB and there should be no effect on the shape change or the stability. 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 [6]. This general idea would suggest that the inviscid approximation should give reliable results for the stability of MEB over a wide range of Reynolds number.

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