

Fission of Multielectron Bubbles in Liquid Helium

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Abstract

The stability of multielectron bubbles (MEBs) in liquid helium is investigated using the liquid-drop model for fissioning nuclei. Whereas a critical positive pressure can make the bubble unstable against fissioning, a small negative pressure suffices to introduce a restoring force preventing any small deformation of the bubble to grow. We also find that there exists an energy barrier making MEBs metastable against fissioning at zero pressure. The results obtained here overcome the difficulties associated with the Rayleigh-Plesset equation previously used to study bubble stability, and shed new light on the limits of achievable bubble geometries in recently proposed experiments devised to stabilize MEBs.

I. INTRODUCTION

Multielectron bubbles (MEBs) are fascinating entities which appear when a surface of bulk helium covered by a 2D film of electrons becomes unstable [1,2]. MEBs are cavities in the liquid helium, filled with electrons that form a 2D spherical layer at the inner surface of the bubble. Recent proposals to stabilize MEBs [3] have stimulated theoretical investigations into its properties [4,5], since this system holds the promise of studying the electron gas in a controlled way, bereft from material impurities. The density of this electron gas can be tuned over more than four orders of magnitude by pressurizing the liquid helium [5] and this tunability would make the observation of a hexatic phase [4] and the quantum melting of a ripplon Wigner lattice [6,7] experimentally feasible. For these investigations, both theoretical and experimental, the question of the stability of multielectron bubbles is of crucial importance. In this paper, the results reported in a recent letter [5] on the pressure dependence of the frequency of the modes of deformation of an MEB are discussed in the framework of new results obtained using the Bohr model for fissioning [8].

The energy and the radius of a multielectron bubble with N electrons can be estimated by balancing the surface tension with the electrostatic Coulomb repulsion [9]. In this approximation, the energy of the MEB is proportional to $N^{4/3}$, so that the energy of a single bubble with N electrons is larger than the energy of two bubbles at infinite distance and with $N/2$ electrons each. The lower energy of the fissioned bubble has led to speculations about the stability of multielectron bubbles. Since MEBs with N up to 10^8 were observed experimentally [1,2], albeit in a transient manner (lasting a few msec), there must exist a formation barrier preventing the fissioning of MEBs. Early investigations ruled out gravitationally induced instabilities and tunneling decay of the bubbles as possible fissioning mechanisms [9]. Salomaa and Williams [10] considered dynamical stability against “boiling off” a single electron from a multielectron bubble and found stability against this type of fission for bubbles with $N > 15 - 20$.

Preceding studies of the small amplitude oscillations of the bubble shape [10,5] have

shown that the quadrupole mode of oscillation has a vanishing frequency when no pressure is applied on the liquid helium. Furthermore it was shown that with increasing positive pressure successive modes can be driven to a vanishing frequency [5]. The amplitude of modes of deformation that have a vanishing frequency can grow until they become of the order of the bubble radius. This deformational instability can lead to fissioning of the bubble. Salomaa and Williams [10] have investigated the dynamics of this deformational instability using coupled Rayleigh-Plesset equations for the deformation amplitudes and the bubble radius, and found that when the initial amplitude of the quadrupole mode of deformation is larger than $\sim 6\%$ of the bubble radius, the amplitude of the oscillation keeps growing as a function of time. However, their method is not valid when the oscillation amplitude becomes comparable to the radius of the bubble; hence we need to develop a new approach to describe the fission process.

II. THE BOHR MODEL FOR FISSIONING MULTIELECTRON BUBBLES

In this paper, we apply the Bohr liquid-drop model of fissioning nuclei [8] to MEBs. This method is valid for both small and large amplitude deformations of atomic nuclei and describes the nucleus as a charged droplet with surface tension. It was recently successfully improved to derive the fragment mass asymmetries in nuclear fission [11]. In such models of nuclear fission, and similar models for the breaking up of homogeneously charged liquid droplets, an approximation is used to calculate the properties of the fission process. This approximation consists of describing the surface of the splitting nucleus (or droplet) in terms of two or three quadratic forms (spheroids and hyperboloids). Such a surface is parametrized in cylindrical coordinates through

$$\rho = \begin{cases} \sqrt{a_L - b_L(z - f_L)^2} & \text{for } z_0 \leq z \leq z_1 \\ \sqrt{a_M - b_M(z - f_M)^2} & \text{for } z_1 \leq z \leq z_2 \\ \sqrt{a_R - b_R(z - f_R)^2} & \text{for } z_2 \leq z \leq z_3 \end{cases} \quad (1)$$

where

a_i is the square of the semimajor axis of the spheroid along the radial direction. For a hyperboloid, a_i is negative ($i = L, M, R$);

b_i is the deformation parameter: the square of radial semimajor axis divided by the square of the longitudinal semimajor axis;

f_i is the centre of the spheroid (hyperboloid);

$z_0 = f_L - a_L/\sqrt{b_L}$ is the leftmost point of the shape;

$z_3 = f_R + a_R/\sqrt{b_R}$ is the rightmost point of the shape.

These parameters are illustrated in Fig. 1. The surface determined by (1) describes the shape of the bubble and allows to investigate both spheroidal bubbles and emerging spheroidal fragments. The shape parameters $\{a_L, a_M, a_R\}$, $\{b_L, b_M, b_R\}$, $\{f_L, f_M, f_R\}$, $\{z_1, z_2\}$ are not independent if one imposes continuity and continuous derivatives at the meeting points of the quadratic forms. These conditions, together with fixing the origin at z_0 to remove translations from the set of shape changes under consideration, allows to eliminate five of the eleven variables. The six independent parameters that are kept in our treatment are: $a_L, b_L, a_R, b_R, f_R - f_L$ and z_1 . For a droplet of incompressible fluid, there would be another constraint (that of fixed volume) to remove one more parameter, but in the case of MEBs the volume does not have to remain constant during deformations [5].

Within this model, an expression for the energy of a bubble with a given shape (fixed by choosing the shape parameters $a_L, b_L, a_R, b_R, f_R - f_L$ and z_1) is obtained. The stable shape of the bubble is found by minimizing the energy as a function of the shape parameters – the shape parameters set up a ‘shape space’. Both the case of the spherical, unsplit bubble and the case of a bubble split in two fragments can be described with appropriate shape parameters, so these two cases can be represented by distinct points in the shape space. The dynamics of the fissioning of the multielectron bubble can then be studied by determining the energy along the trajectories in shape space which go from the point representing the spherical, unsplit bubble to the point representing a bubble split in two. The unsplit bubble

may have a higher energy than the fissioned bubble, but if there is an energy barrier along each trajectory in shape space, then the single bubble is metastable with respect to fissioning. The height of the energy barrier along the optimal fissioning trajectory is the metastability energy barrier.

In section III we set up the expression for the energy of a bubble with given shape parameters (the energy of a given point in shape space). In section IV, we discuss the results of the minimization of the energy in shape space and the results for the optimal trajectory for fissioning of a multielectron bubble.

III. ENERGY OF A DEFORMED MEB

The energy of an MEB is determined as a function of the shape of the bubble by three contributions: (i) the surface tension energy $E_\sigma = \sigma S$ where $\sigma = 3.6 \times 10^{-4}$ J/m² is the surface tension of liquid helium and S is the bubble surface, (ii) the pressure-related energy $E_p = pV$ where p is the (experimentally tunable) difference in pressure inside and outside the bubble and V is the volume of the MEB; and (iii) the electrostatic repulsion energy E_C of the electrons. The first two terms are easily evaluated since the surface and volume of the bubble are related straightforwardly to the shape parameters. The surface is given by

$$S = \sum_{i=1,2,3 \equiv L,M,R} \pi \int_{z_{i-1}-f_i}^{z_i-f_i} \sqrt{a_i + b_i(b_i - 1)x^2} dx, \quad (2)$$

where in the case of split bubbles ($b_M < 0$ and $a_M < 0$) the integration domain should not include the region of space in between the bubble fragments. The volume is

$$V = \int_{f_L - \sqrt{a_L/b_L}}^{f_R + \sqrt{a_R/b_R}} dz \pi \rho^2(z), \quad (3)$$

where the cylindrical radius $\rho(z)$ is given by (1). The integral in expression (3) is a piecewise sum of integrals of the type

$$\int_{z_a}^{z_b} \pi [a + b(z - f)^2] dz = \pi a(z_b - z_a) - \frac{b}{3}((z_b - f)^3 - (z_a - f)^3). \quad (4)$$

The evaluation of the electrostatic energy is greatly simplified by the observation [12] that the quantum mechanical correction (the exchange term) is negligible for the determination of the total electrostatic energy. Furthermore the electrons in the bubble are not smeared out throughout the bubble volume, but remain in a nanometer thin, effectively two-dimensional layer anchored to the helium surface [9,10]. This layer conforms to the helium surface also when the bubble deforms [10]. We will assume that the surface density of electrons is homogeneous along the surface and equal to $n_s = N/S$ where N is the number of electrons and S is the area of the deformed bubble. Some justification of this assumption comes from a recent calculation of the coupled ripplon-phonon modes of oscillation of an MEB [6]. In the calculation of Ref. [6], the coupling between the modes of deformation of the helium surface (“rippbons”) and the redistribution of the surface density of electrons (“phonons”) was investigated, and it was derived that this coupling was weak enough not to affect strongly the oscillation frequencies of ripples and phonons. Within the assumptions described in this paragraph, we may write the electrostatic repulsion energy as

$$E_C = \int d^3\mathbf{r} \int d^3\mathbf{r}' \frac{(n_s e) \cdot (n_s e)}{\varepsilon |\mathbf{r} - \mathbf{r}'|} \quad (5)$$

$$= \frac{(2\pi n_s e)^2}{\varepsilon} \int_{z_0}^{z_3} dz \int_{z_0}^{z_3} dz' \frac{2}{\pi} \int_0^{\infty} dk \cos[k(z - z')] I_0(k\rho_{<}) K_0(k\rho_{>}) \rho(z) \rho(z'), \quad (6)$$

where $\rho_{<} = \min[\rho(z), \rho(z')]$ is the smallest of the two cylindrical radii and $\rho_{>} = \max[\rho(z), \rho(z')]$ is the largest. I_0 is the modified Bessel function of the first kind with index 0, and K_0 is the modified Bessel function of the second kind with index 0.

The total energy of a deformed bubble with given shape parameters is then given by

$$E = \sigma S + pV + E_C. \quad (7)$$

Expressions (2),(3), and (5) allow to calculate the total energy E of the bubble for any point in the shape space discussed in the previous section. In Ref. [5] the energy of an

MEB undergoing small-amplitude oscillations was calculated. The total energy (7) of the deformed bubble used in the present treatment agrees perfectly with the results of Ref. [5] in those cases where both approaches are applicable. Also the equilibrium radius of a spherical bubble, obtained by minimizing the expression (7) with respect to the radius, obviously agrees with the result of [5].

IV. RESULTS AND DISCUSSIONS

To describe the fissioning of the bubble through a shape deformation, we will investigate the trajectory in shape space that has the lowest energy and that starts from the spherical bubble with equilibrium diameter $d = z_3 - z_0 = 2R_b$. The equilibrium radius R_b for the spherical bubble with N electrons can be found by minimizing

$$E^{\text{spherical}} = 4\pi\sigma R_b^2 + \frac{4\pi}{3}pR_b^3 + \frac{N^2e^2}{2\epsilon R_b}. \quad (8)$$

When $p = 0$, this radius is $R_b = \sqrt[3]{N^2e^2/(8\pi\epsilon\sigma)}$ (for example, with $N = 10^4$ electrons in the bubble, $R_b = 1.064 \mu\text{m}$). To investigate the presence of an energy barrier stabilizing the MEB against fissioning, we calculate the minimum energy (and shape) of a bubble as a function of $d = z_3 - z_0$, the elongation of the shape along the axis of symmetry (see Fig. 1).

A. Zero pressure

The results of this minimization are shown in Fig. 2, for an MEB with $N = 10^4$ electrons at $p = 0$. For every given value of d (the x-axis), the shape parameters $a_L, b_L, a_R, b_R, f_R - f_L, z_1$ are varied under the constraint $z_3 - z_0 = d$. The optimal variational energy per electron E/N given by expression (7) is shown in Fig. 2, along with the optimal variational shape for some selected points (marked by A,B,E).

At $d = 2R_b$, we find that the optimal shape is a spherical bubble (the point marked by A in Fig. 1). At increased d , the optimal bubble shape becomes ellipsoidal (for example,

point B). The energy as a function of d , for $d < 3.012 \mu\text{m}$, is independent of d . This is in agreement with previous results on the frequency of the vibrational modes of the bubble [10,5]. The eigenmodes of vibration of the bubble surface are characterized by spherical harmonic mode numbers $\{\ell, m\}$, and the eigenfrequencies (at $p = 0$) are given by [10,5]:

$$\omega(\ell) = \sqrt{\frac{\sigma}{\rho R_b^3}(\ell - 2)(\ell^2 - 1)} \quad (9)$$

The deformation corresponding to point B in Fig. 2 is an $\ell = 2$ eigenmode of the system. This mode has zero frequency according to expression (9). This means that it does not cost energy to introduce small amplitude $\ell = 2$ deformations of the bubble, and the energy should remain constant as a function of d , as it does (see Fig. 2).

The curve representing the minimum variational energy as a function of d has a sudden change of slope near $d = 3.012 \mu\text{m}$. The inset shows more results near this point. We found that near this point two different minima exist, corresponding to two topologically distinct shapes. On the horizontal part of the curve (dashed line containing point B or point C in the inset of Fig. 2) the shape is an ellipsoid. On the curve with negative slope (full curve containing point E or point D in the inset of Fig. 2), the optimal shape is a bubble split up into two fragments with $N/2$ electrons in each of the fission fragments. These two topologies compete for the global minimum. For $d > 3.012 \mu\text{m}$, the fissioned bubble has lower energy: point D is lower in energy than point C.

Let's investigate whether there exists some energy barrier stabilizing the elongated bubble of point C against fission towards the fissioned shape corresponding to D. To split up an elongated bubble (C) into fission fragments (D), the bubble shape has to deform through intermediate shapes, as those shown in Fig. 3. These intermediate shapes appearing during the fission trace out a trajectory in shape space, starting at the point corresponding to C and ending in the point corresponding to D. This trajectory can be parametrized by an interpolation parameter which is zero at the starting point (C) and 1 at the endpoint (D). The energy of the intermediate shape as a function of the interpolation parameter is shown in Fig. 3. It is clear that the two minima (elongated bubble, interpolation parameter=0,

and split-up bubble, interpolation parameter=1) are separated by an energy barrier, of the order of 0.2 eV per electron. During the process of fission, the elongated bubble has to be deformed to create the neck between the fission fragment and the parent bubble. This deformation must involve high- ℓ modes of deformations, which cost energy $\omega(\ell) > 0$. This gives rise to the energy barrier shown in Fig. 3.

At this point it is also possible to clarify why the main limitation of the Bohr model of fissioning does not affect our result. This limitation is that only the splitting off of a single fragment -albeit of any size- can be described. A fissioning process whereby the bubble splits in three or more parts cannot be modelled realistically using only three quadratic surfaces. Nevertheless it is clear that such a fissioning process would involve high- ℓ modes of deformation: the more fragments appear at the surface, the more complicatedly it has to be deformed. Such a fissioning process would therefore require even higher energy and its realization would be much less likely. Note that the size of the fission fragments ($N/2$) is in agreement with expression (8) for the energy of a spherical bubble, $E^{\text{spherical}}(N) \propto N^{4/3}$. This can be easily shown from minimizing $E^{\text{spherical}}(m) + E^{\text{spherical}}(N - m)$ with respect to the optimal fragment size m .

B. Effect of positive pressure

In Ref. [5], it was shown that with increasing pressure, more and more eigenmodes of the bubble deformation obtain vanishing frequencies. The pressure dependence of the frequency of these modes is given by [5]:

$$\omega(\ell) = \sqrt{\frac{\ell + 1}{\rho R_b^3} \left[\sigma(\ell^2 + \ell + 2) + 2pR_b - \frac{N^2 e^2}{4\pi\epsilon R_b^3} \ell \right]} \quad (10)$$

where R_b is now the equilibrium radius under pressure, which satisfies $2pR_b + 4\sigma = e^2 N^2 / (4\pi\epsilon R_b^3)$. The effect of positive pressure is such that some modes of deformation become ‘soft’ (i.e. have vanishing frequency): for example, for a bubble with 10^4 electrons at $p = 3$ mbar ($R_b = 0.95 \mu\text{m}$) the $\ell = 2$ and $\ell = 3$ modes of deformation are unstable [5].

Fig. 4 shows the result for the minimum variational energy of a bubble with 10^4 electrons at $p = 3$ mbar ($R_b = 0.95 \mu\text{m}$) as a function of the elongation d . As the bubble deforms (along the curve containing points ABC in Fig. 4), its energy decreases. Also, in contrast with the zero-pressure case, the lowest energy shape for $d < 2.68 \mu\text{m}$ exhibits deformations with both $\ell = 2$ and $\ell = 3$ character. This is in agreement with (10).

Again two bubble topologies compete for the global minimum: the solution where the shape is a singly connected surface (squares in Fig. 4), and the solution where the bubble is split in two fragments (circles in Fig. 4). For $d < 2.68 \mu\text{m}$ the singly connected, unsplit bubble has the lower energy, whereas at larger d , the fissioned bubble has lower energy.

The availability of higher angular momentum modes (such as $\ell = 3$) allows the MEB to deform to create a ‘neck’ connecting an emergent fission fragment with the ‘parent’ bubble, without increasing the total energy of the bubble. In the inset of Fig. 4, we show the variational energy of the intermediate shapes assumed by the bubble during a fission process going from point C to point D in Fig. 4. The ‘interpolation parameter’ traces out the trajectory in shape space during the fission process, as described in the discussion of Fig. 2 in the previous subsection. The energy barrier which was present in the case of zero pressure is no longer present, and we conclude that pressurized MEBs are unstable when the pressure is high enough to drive $\ell > 2$ modes unstable.

In ref. [5] it was shown that the pressure at which a particular mode of deformation becomes unstable, is larger for bubbles with fewer electrons. Thus, when the pressure is raised so as to make a bubble with N electrons unstable and fission that bubble, the resulting fission products with $N/2$ electrons may still be metastable. To fission also those fragments the pressure needs to be raised further.

C. Effect of negative pressure

Liquid helium can sustain a negative pressure of -9 bar (± 1 bar) before it cavitates [13]. Here, a negative pressure on the MEB means that the force associated with this pressure

is directed outward, away from the bubble center. Shikin [9] estimated in a simplified model that MEBs with a radius larger than the zero-pressure equilibrium radius $R_b(p = 0)$ may be metastable in the sense that there exists a restoring force which counteracts small deformations. Salomaa and Williams [10] found that the nonlinear coupling between the radial mode of oscillation and the deformational modes of oscillation may lead to a small increase in the time-averaged radius, stabilizing the bubble. A straightforward way to increase the bubble radius is by applying negative pressure. In Ref. [5] we showed that for negative pressures all the eigenmodes of deformation acquire a positive frequency.

The liquid-drop model of fission allows to describe deformations beyond the small-amplitude region of validity of the Rayleigh-Plesset equations used in [10]. Figure 5 shows the result of negative pressures on the shape and fissioning of the bubble. The variational minimum energy is again shown as a function of the elongation along the axis of symmetry $d = z_3 - z_0$, for a bubble with $N = 10^4$ electrons at $p = -3$ mbar ($R_b = 1.5 \mu\text{m}$). In contrast to unpressurized MEBs and MEBs at positive pressure, now three geometries compete for the global minimum instead of two: (A) an $\ell = 2$ deformed bubble, (B) the split-up bubble, and (C) a spherical bubble with expanded radius. To deform the bubble from any of these three shapes into another, intermediate shapes have to be assumed and an energy barrier will be present like in the case of zero pressure.

But, apart from this energy barrier, another energy barrier is present: from the increase of the energy with increasing d in Fig. 5, it is clear that there is a restoring force for small-amplitude deformations. This restoring force was absent for $p = 0$ (compare Fig. 5 with Fig. 2) and in the case of $p > 0$ the force was of opposite sign and driving the instability (compare Fig. 5 with Fig. 4). The restoring force is related to the lowest frequency of the eigenmodes, $\omega(\ell = 2)$ which becomes positive at negative pressures, as derived in [5]. The restoring force results in an additional energy barrier of 0.15 eV per electron in the case studied here. We emphasize that the additional energy barrier related to the intermediate bubble shapes (as illustrated by Fig. 3 for the $p = 0$ case) is present also at negative pressures (but not at positive pressures large enough to drive $\ell > 2$ modes to zero frequency).

Note furthermore that, if there is a driving force which can excite the deformation to large amplitude and overcome the barriers, the MEB may be destroyed in one of two ways: either by fissioning of the bubble, after which the fission fragments move away from each other, or the MEB may keep expanding until it fills a volume large enough to counteract the negative pressure (i.e. the MEB serves as a nucleation center for cavitation).

V. CONCLUSIONS

In this paper, the liquid drop model of fissioning was applied to the problem of multielectron bubbles in liquid helium. We found that, even though there exists, at $p = 0$, a mode of deformation which can grow without increasing the total energy of the bubble, there is still an energy barrier present which prevents fissioning of the bubble. This barrier was explained by the intermediate shapes that the fissioning bubble has to assume in order to create a neck between the emerging fission fragment and the parent bubble. These intermediate shapes involve eigenmodes of deformation which cost substantial energy (0.2 eV per electron for a 10000 electron bubble). At positive pressure, these higher angular momentum modes can obtain a vanishing frequency as discussed in [5], and this causes the energy barrier to vanish and the bubble to become unstable. However, at negative pressure, when all the modes of deformation have a non-vanishing frequency, there is an additional element of stability in that there is a restoring force which counteracts small amplitude deformations. The present study, based on the liquid drop model, independently confirms and extends the conclusions presented in a previous letter [5], namely that a positive pressure can make the MEB unstable, whereas a small negative pressure makes the MEB metastable against fission. The additional result presented in this paper is that also at zero pressure the MEB is metastable.

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FIGURE CAPTIONS

FIG. 1. The model shape for a fissioning MEB consists of three quadratic forms, for example two spheroids connected by a hyperboloidal neck as illustrated in this figure. The relation between the model shape and the parameters used in the text, expression (1), is shown in this figure.

FIG. 2. The minimum variational energy per electron (in eV) of an $N = 10^4$ electron MEB at $p = 0$ is shown as a function of the bubble elongation d (microns). This energy is obtained by minimizing expression (7) with respect to the shape parameters illustrated in Fig. 1, and subject to the constraint $z_3 - z_0 = d$. The symbols (squares and diamonds) are the results of the minimization and the curves are guides for the eye. For some points of interest (A,B,E) the corresponding shape of the bubble is illustrated. Two bubble shapes compete for the minimum energy: an elliptically elongated bubble (points on the dashed curve) and the fissioned bubble (points on the full curve). For $d < 3.012 \mu\text{m}$ the elongated bubble is the minimum energy shape whereas for larger d the fissioned bubble is the minimum energy shape.

FIG. 3. When a 10^4 electron MEB at zero pressure is elongated more than $d = 3.012 \mu\text{m}$, the minimization of the total energy shows that it becomes energetically favorable to split the bubble in two equal fragments. However, in order to fission the elongated bubble, it has to deform in such a way that a neck develops where the bubble can split in two. These intermediate shapes of the bubble are higher in energy than either the elongated shape (C) or the split bubble (D), and give rise to the presence of an energy barrier. Points C and D in this figure correspond to those of Fig. 2; the fission process traces out a trajectory in shape space connecting C and D and parametrized by an interpolation parameter ranging from 0 (C) to 1 (D).

FIG. 4. The minimum variational energy per electron of an $N = 10^4$ electron MEB at $p = 3 \text{ mbar}$ is shown as a function of the bubble elongation d (in micron). For some points

of interest (A,B,C,D), the bubble shape is illustrated. In the inset, the energy is shown as a function of the interpolation parameter which takes the shape parameters from point C into those from point D. Comparing this figure with Figs. 2,3, it is clear that the bubble now can decrease its energy monotonically while splitting: applying a positive pressure so that $\ell > 2$ modes of deformation obtain a vanishing frequency can remove the energy barrier and make the MEB unstable.

FIG. 5. The minimum variational energy per electron of an $N = 10^4$ electron MEB at $p = -3$ mbar is shown as a function of the bubble elongation $d = z_3 - z_0$ (in micron). Three geometries compete for the global minimum: the $\ell = 2$ mode of deformation of the bubble (point A), the split bubble (point B) and the spherical bubble with large radius (point C). At negative pressure, there exists an additional energy barrier associated with a restoring force which prevents small-amplitude deformations ($d < 4 \mu\text{m}$) from growing. The inset shows the region near the maximum of the energy barrier in more detail.

Figure 1

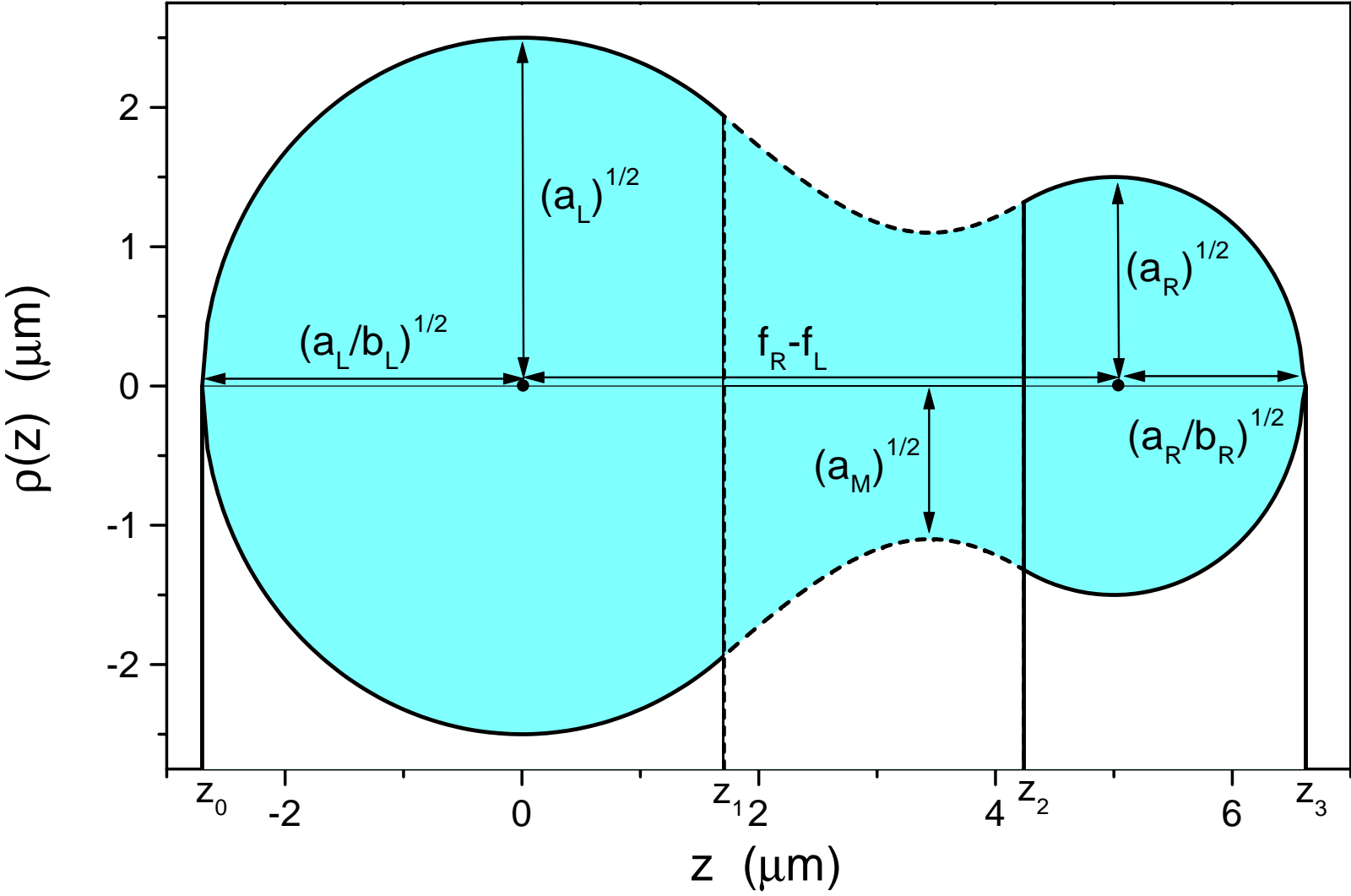


Figure 1

Figure 2

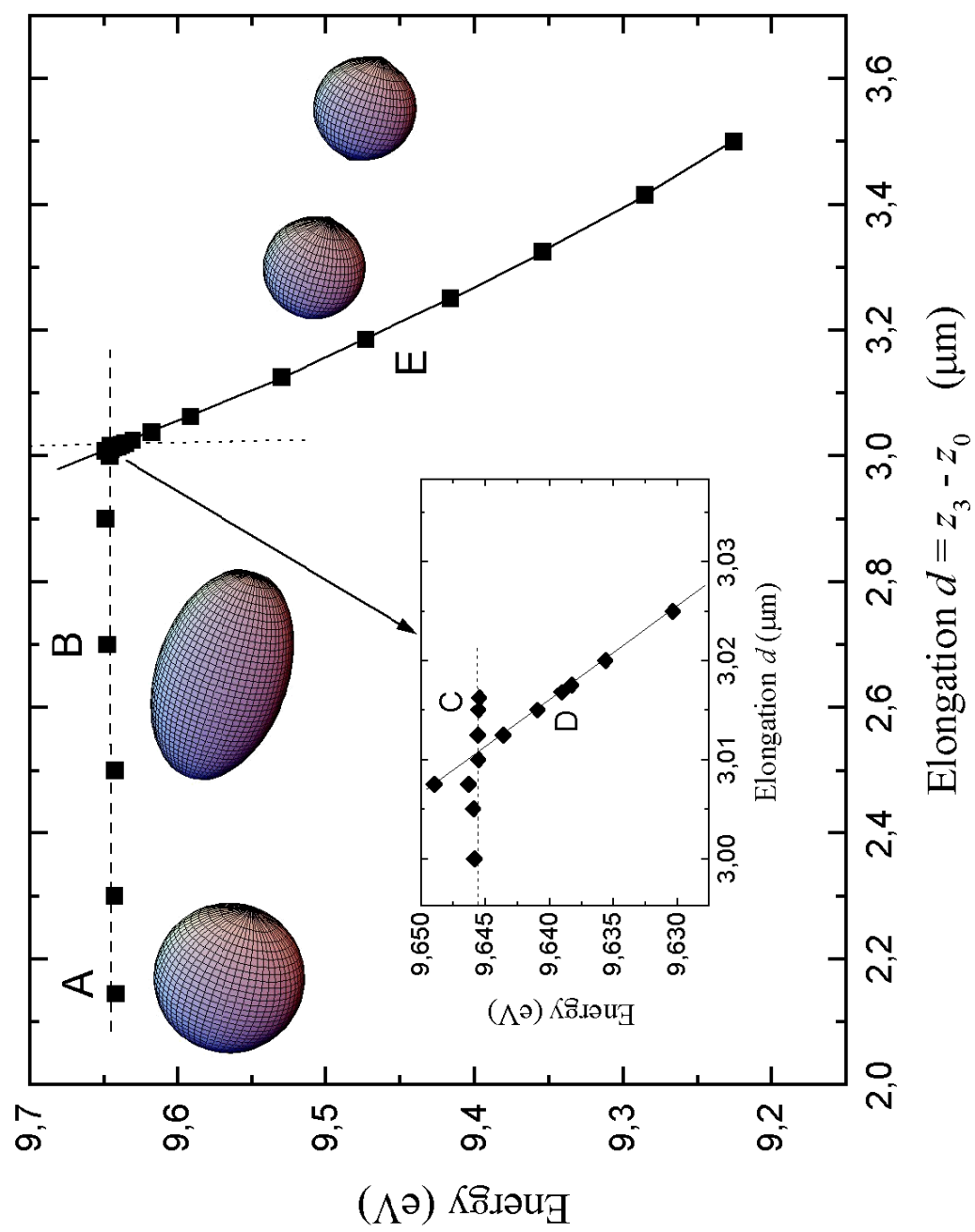


Figure 2

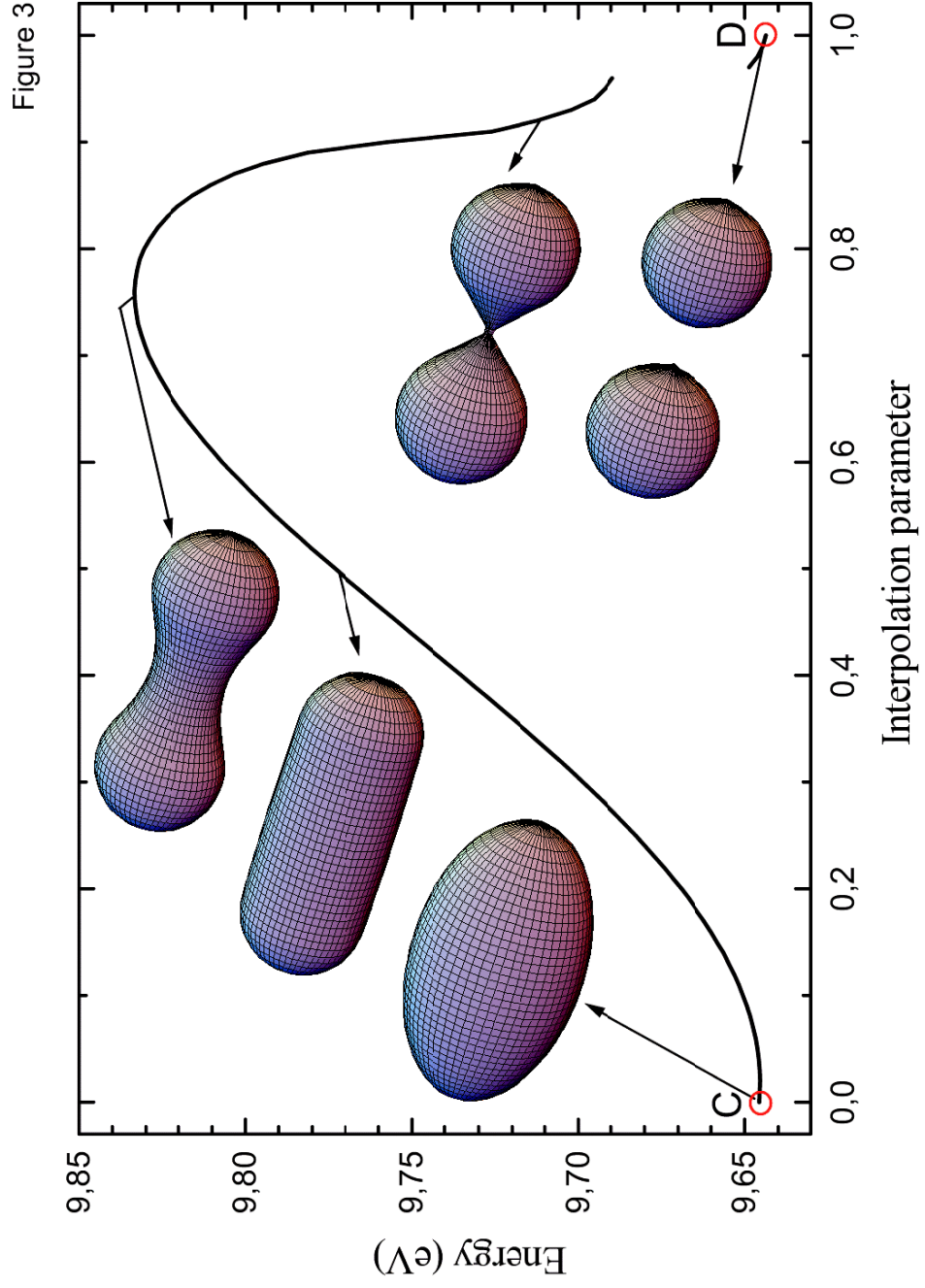


Figure 3

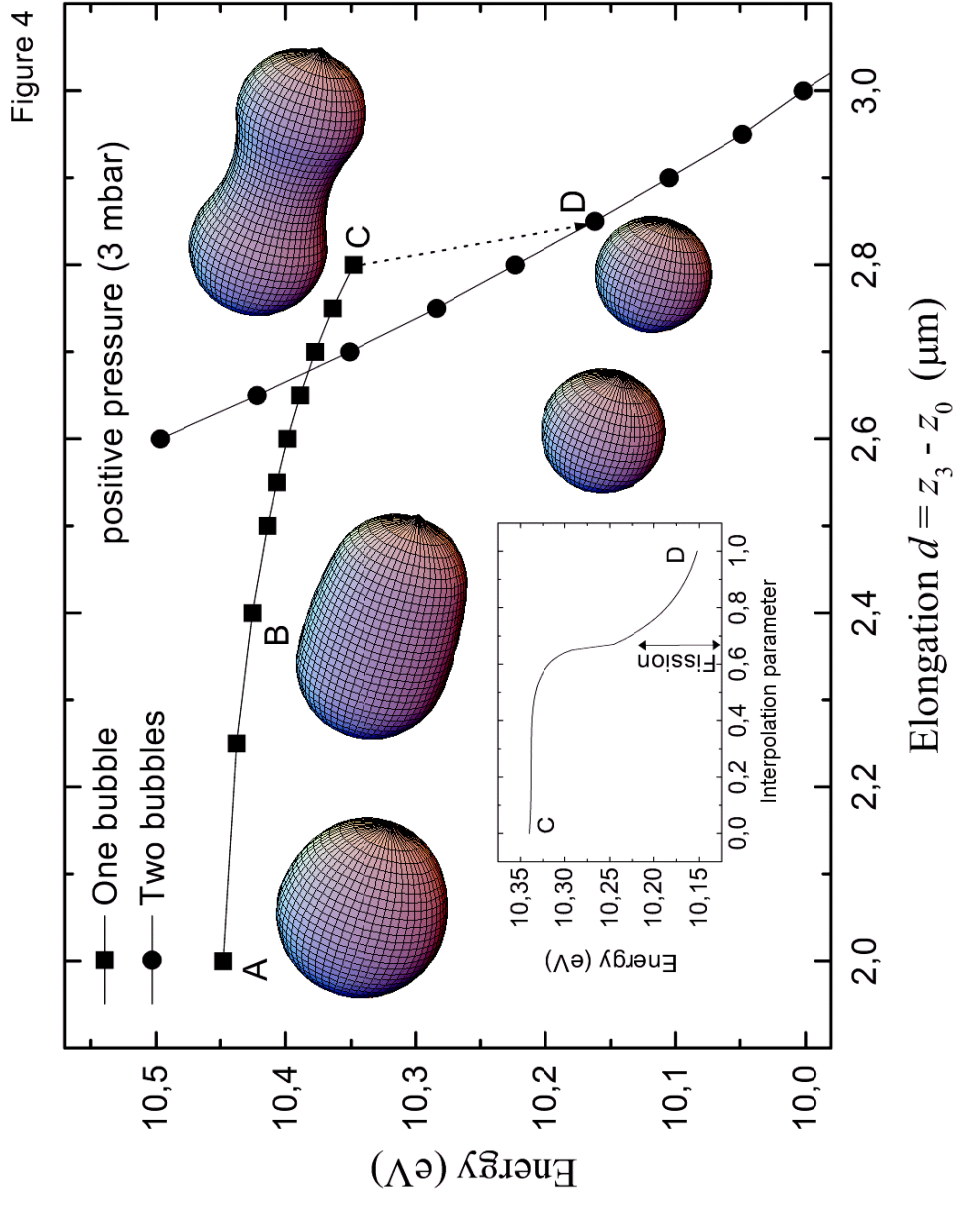


Figure 4

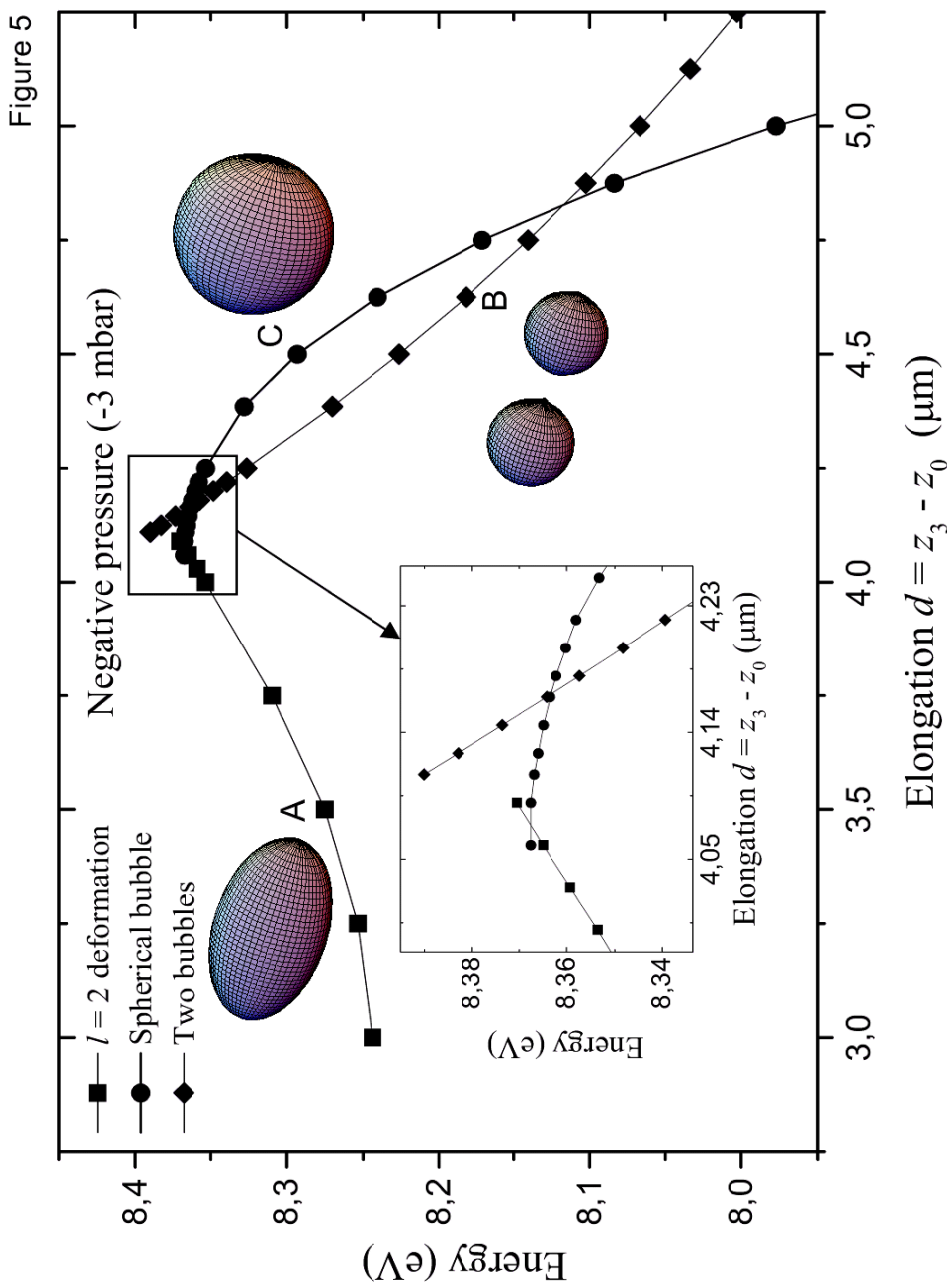


Figure 5

Figure 5