Coupled Ripplon-Plasmon Modes in a Multielectron Bubble

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Abstract

In multielectron bubbles, the electrons form an effectively two-dimensional layer at the inner surface of the bubble in helium. The modes of oscillation of the bubble surface (the ripplons) are influenced by the charge redistribution of the electrons along the surface. The dispersion relation for these charge redistribution modes (‘longitudinal plasmons’) is derived and the coupling of these modes to the ripplons is analysed. We find that the ripplon-plasmon coupling in a multielectron bubble differs markedly from that of electrons at a flat helium surface. An equation is presented relating the spherical harmonic components of the charge redistribution to those of the shape deformation of the bubble.

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I. INTRODUCTION

Multielectron bubbles are 0.1 \( \mu \text{m} \) – 100 \( \mu \text{m} \) sized cavities inside liquid helium, that contain helium vapor at vapor pressure and a nanometer-thick electron layer anchored to the surface of the bubble \[1\]. The dependence of the radius of the bubble on the pressure applied to the bubble \[2\] allows to vary the surface density of the spherical two-dimensional electron system in the bubble over nearly four orders of magnitude. As such, these objects offer the prospect to study the two-dimensional electron system on helium in regimes hitherto inaccessible. In particular, this would allow to study the two-dimensional electron Wigner crystal at densities not currently achievable for electrons on a flat helium surface.

Early experimental evidence for 2D Wigner crystallization of electrons on a liquid-He surface \[3\] relied on the detection of the coupled plasmon-ripplon modes \[4\]. In Ref. \[2\], the ripplon modes characteristic for a bubble inside liquid helium were calculated and investigated as a function of the pressure applied on the bubble. In this communication we extend this investigation and take into account the redistribution of charge along the bubble surface when the bubble deforms. This allows us to derive the dispersion relation for the spherical ‘plasmon’ modes and the plasmon-ripplon coupling peculiar for the spherical surface.

II. SMALL AMPLITUDE DEFORMATIONS AND CHARGE REDISTRIBUTION

The surface of a bubble in liquid helium can be described by a function \( R(\Omega) \) which gives the distance of the helium surface from the center of the bubble, in a direction given by the spherical angle \( \Omega = \{\theta, \phi\} \). In equilibrium (at zero pressure) the multielectron bubble has a spherical surface with radius \( R_b \). We expand the deformation away from the spherical surface in spherical harmonics:

\[
R(\Omega) = R_b + \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} Q_{\ell m} Y_{\ell m}(\Omega),
\]

(1)

where \( R_b \) is the angle-averaged radius of the bubble, \( Q_{\ell m} \) is the amplitude of the spherical harmonic deformation mode indexed by \( \{\ell, m\} \), and \( Y_{\ell m}(\Omega) \) is the corresponding spherical harmonic. These modes of deformation are referred to as ripplons in analogy with the surface modes of a flat surface of liquid helium. In Ref. \[2\] the dispersion relation for the ripplons in a multielectron bubble has been studied as a function of pressure and related to the bubble stability. In this communication, we extend the results of \[2\] in order to take the coupling and mixing of the ripplons and the electronic modes into account.

In a multielectron bubble, the electrons are confined to a thin (1 nm) layer at the inner surface of the bubble. This layer is anchored to the surface of the bubble, so that when the surface deforms, the layer conforms to the new bubble surface. The electrons can redistribute themselves inside the spherical layer, so that the surface density of electrons is no longer uniform. We describe the surface density with the function \( n_S(\Omega) \) giving the number of electrons in a solid angle \( d\Omega \) around \( \Omega \):

\[
n_S(\Omega) = \frac{N}{4\pi R_b^2} + \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} n_{\ell m} Y_{\ell m}(\Omega).
\]

(2)
This function is normalized such that \( N = \int n_s(\Omega) d\Omega \) is the total number of electrons in the bubble or on the droplet. The \( n_{\ell m} \)‘s represent the amplitudes of charge redistributions corresponding to spherical harmonics \( Y_{\ell m} \).

The total potential energy of the multielectron bubble or droplet can be separated into several contributions: (i) a term from the surface tension energy \( U_S = \sigma S \), with \( \sigma \approx 3.6 \times 10^{-4} \) J/m\(^2\) and \( S \) the surface of the deformed bubble or droplet; (ii) a pressure related term \( U_p = pV \), with \( p \) the difference in pressure outside and inside the bubble and \( V \) the volume of the bubble; (iii) a term representing the electrostatic energy \( U_C \) of the electron layer. In this last term, the quantum corrections (such as the exchange contribution) to the electrostatic energy can be neglected \[5\]. The first two terms, \( U_S \) and \( U_p \), have been derived in \[2\]:

\[
U_S = 4\pi \sigma R_b^2 + \frac{\sigma}{2} \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} (\ell^2 + \ell + 2) |Q_{\ell m}|^2, \quad (3)
\]

\[
U_p = \frac{4\pi}{3} pR_b^3 + pR_b \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} |Q_{\ell m}|^2. \quad (4)
\]

Whereas \( U_S \) is an exact expression, \( U_p \) is an expansion up to second order in \( Q_{\ell m} \) and the third order term has been neglected. In what follows, we assume small amplitude deformations and small amplitude charge redistributions such that

\[
\sqrt{\ell(\ell+1)} |Q_{\ell m}| \ll R_b, \quad (5)
\]

\[
\sqrt{\ell(\ell+1)} |n_{\ell m}| \ll \frac{N}{4\pi R_b^2}. \quad (6)
\]

The electrostatic potential \( V(\mathbf{r}) \) of the deformed MEB with a non-uniform surface electron density is calculated straightforwardly within the framework of the Maxwell equations by expanding the potential inside and outside the deformed bubble in their respective spherical decompositions and imposing the electrostatic boundary conditions at the surface. The potential energy associated with this electrostatic potential is given by

\[
U_C = -(1/2) \int n_s(\Omega) V(R, \Omega) d\Omega. \quad (7)
\]

If we keep only terms up to second order in both \( n_{\ell m} \) and \( Q_{\ell m} \), we find

\[
U_C = \frac{e^2 N^2}{2\varepsilon R_b} + 2\pi e^2 R_b^3 \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{|n_{\ell m}|^2}{\varepsilon_1 \ell + \varepsilon_2 (\ell + 1)}
\]

\[
- \frac{e^2 N^2}{8\pi \varepsilon R_b^3} \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{\ell + 1}{\varepsilon_1 \ell + \varepsilon_2 (\ell + 1)} |Q_{\ell m}|^2
\]

\[
- e^2 N \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{\ell + 1}{\varepsilon_1 \ell + \varepsilon_2 (\ell + 1)} n_{\ell m} Q_{\ell m}^* . \quad (8)
\]

where \( e \) is the electron charge, \( \varepsilon_1 \) is the dielectric constant of the medium inside the surface, and \( \varepsilon_2 \) is the dielectric constant of the medium outside the surface. For a multielectron
bubble, $\varepsilon_2 = \varepsilon$ with $\varepsilon = 1.0572$ the dielectric constant of liquid helium, and $\varepsilon_1 = 1$ (if the vapour pressure of the helium is low enough, the bubble has vacuum inside).

The ripplon contribution to the kinetic energy of the MEB is associated with the motion of the liquid helium surface. Following a derivation of Lord Rayleigh for oscillating liquid droplets [2], we find

$$T_r = \frac{\rho R_b^3}{2} \ddot{R}_b^2 + \frac{\rho R_b^3}{2} \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{1}{\ell + 1} |\dot{Q}_{\ell m}|^2 .$$

(9)

where $\rho \approx 145 \text{ kg/m}^3$ is the density of liquid helium. Note that for a bubble, $\ell + 1$ appears in the denominator instead of $\ell$ for the droplet in Lord Rayleigh’s treatment.

The ‘classical’ kinetic energy of the electrons is given by

$$T_e = \frac{m_e}{2} \sum_{j=1}^{N} \dot{\mathbf{w}}^2(\mathbf{r}_j)$$

(10)

with $m_e$ the electron mass, and $\mathbf{w}(\mathbf{r}_j)$ represents the displacement of electron $j$ out of its equilibrium position $\mathbf{r}_j$. Since we assumed [3], we can use the formula

$$T_e = \frac{N m_e}{8\pi} \int_{\text{surface}} \dot{\mathbf{w}}^2(\mathbf{r})d^2\mathbf{r}$$

(11)

and express the field of displacements $\mathbf{w}(\mathbf{r})$ as a sum of a longitudinal field $\mathbf{w}_L(\mathbf{r})$ and transverse field $\mathbf{w}_T(\mathbf{r})$. We investigate the effect of the longitudinal field, which can be written as a gradient of a scalar potential. The (divergence-free) transverse field is not considered here since it does not couple to the ripplons. Using

$$\nabla \cdot \mathbf{w}_L(\mathbf{r}) = 1 - \frac{4\pi R_b^2}{N} n_S(\theta, \phi)$$

(12)

we can express the kinetic energy of the electrons as

$$T_e = \frac{1}{2} \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{4\pi m_e R_b^6}{N\ell(\ell + 1)} |\dot{n}_{\ell m}|^2$$

(13)

We have checked that our approach yielded, for the 3D and the 2D electron gas, the known expressions for the plasma frequencies.

III. RESULTS AND DISCUSSION

The full Lagrangian of the bubble, including the ripplon modes and charge redistribution modes, is given by substituting expressions (9), (13), [3], [4], [5] in $\mathcal{L} = T_r + T_e - U_S - U_p - U_C$. The result can be brought in the following form

$$\mathcal{L} = \mathcal{L}_R + \sum_{\ell,m} \frac{M_e}{2} \left( \dot{Q}_{\ell m}^2 - \omega_r^2(\ell) Q_{\ell m}^2 \right)$$

$$+ \sum_{\ell,m} \frac{m_e}{2} \left( \dot{n}_{\ell m}^2 - \omega_p^2(\ell) n_{\ell m}^2 \right) + \sum_{\ell,m} \gamma_{\ell m} \dot{n}_{\ell m} Q_{\ell m}.$$
This is the central result of this communication. We will now proceed to discuss the result term by term. The first term in expression (14) contains the Lagrangian describing the radial motion (the anharmonic breathing mode):

$$\mathcal{L}_R = \frac{\rho R_b^3}{2} \dot{R}_b^2 - \frac{4\pi}{3} p R_b^3 - 4\pi \sigma R_b^2 - \frac{N^2 e^2}{2\varepsilon R_b^3}. \tag{15}$$

In what follows we assume that $\dot{R}_b = 0$ and the bubble radius is given by its equilibrium value.

### A. Ripplon modes

The Lagrangian $\mathcal{L}$ also contains a part representing the harmonic oscillation of the ripplon modes, with oscillator mass term

$$M_\ell = \frac{\rho R_b^3}{\ell + 1}, \tag{16}$$

and bare ripplon frequency:

$$\omega_\ell(\ell) = \sqrt{\frac{1}{M_\ell} \left[ \sigma (\ell^2 + \ell + 1) + p R_b - \frac{N^2 e^2}{4\pi \varepsilon R_b^3} \frac{\ell^2 - \varepsilon (\ell + 1)}{\ell + \varepsilon (\ell + 1)} \right]} \tag{17}$$

The bare ripplon frequencies and their dependence on the pressure were the subject of [2]. Note that, in the limit of very large bubbles, $R_b \to \infty$, the dispersion relation for ripplons on a flat helium surface [7] is recovered: if the momentum $k$ is identified with $\ell/R_b$, then the typical $k^{3/2}$ is retrieved. Note furthermore that, if $R_b$ is set equal to the equilibrium coulomb radius, the modes $\ell = 1, 2$ obtain a zero frequency in agreement with the result of Salomaa and Williams [8]. For typical multielectron bubbles with radii in the range of 1-100 $\mu$m (and numbers of electrons of the order $10^4 - 10^7$) the ripplon frequencies for $\ell < 1000$ lie typically in the MHz-GHz range.

### B. Longitudinal plasmon modes

Next, the Lagrangean (14) contains a part representing the (small amplitude) harmonic oscillation of the classical charge redistribution around its equilibrium. The oscillator mass term for these oscillations is

$$m_\ell = \frac{4\pi m_e R_b^6}{N\ell(\ell + 1)}. \tag{18}$$

and the corresponding frequencies are given by

$$\omega_\ell(\ell) = \sqrt{\frac{N e^2}{m_e R_b^6} \frac{\ell(\ell + 1)}{\ell + \varepsilon (\ell + 1)}}. \tag{19}$$

In the limit of large bubbles, this frequency corresponds to that of the longitudinal plasmon frequency of a 2D electron system on the helium surface [9]. If the electrons form a Wigner lattice, this is the longitudinal phonon frequency. If the momentum $k$ is identified with $\ell/R_b$, then the typical $k^{1/2}$ dispersions for this longitudinal plasmon is retrieved. Based on this
correspondence, we will refer to these modes in a multielectron bubble as the ‘(longitudinal) plasmon modes of the MEB’ (or ‘longitudinal phonon modes of the MEB’ if the electrons form a Wigner lattice).

For typical multielectron bubbles with radii in the range of 1-100 µm (and numbers of electrons of the order $10^4-10^7$) the longitudinal plasmon frequencies for $\ell < 1000$ lie typically in the GHz-THz range. For bubbles with $N > 10^5$, the longitudinal plasmon frequencies are larger than the ripplon frequencies, $\omega_p(\ell) \gg \omega_r(\ell)$.

The equations of motion for the electron charge redistribution on a deformed bubble are given by:

$$\frac{d}{dt} \frac{\partial L}{\partial \dot{n}_{\ell m}} = \frac{\partial L}{\partial n_{\ell m}} \iff m_\ell \ddot{n}_{\ell m} = -m_\ell \omega_p^2(\ell) n_{\ell m} + \gamma_\ell Q_{\ell m}. \quad (20)$$

When $\omega_p(\ell) \gg \omega_r(\ell)$ the electrons can redistribute much faster on the surface of the bubble than the bubble surface can deform. Thus, we can make an adiabatic approximation to find the charge redistribution of the electrons on a bubble with a given deformation. We find from (20) that if the bubble surface deformation is described by a given set of spherical deformation amplitudes $Q_{\ell m}$, the equilibrium charge distribution on the deformed bubble must satisfy the relation

$$\frac{n_{\ell m}}{n_0} = \frac{(\ell + 1) Q_{\ell m}}{2 R_b^2}, \quad (21)$$

with $n_0 = N/(4\pi R_b^2)$.

C. Ripplon - longitudinal plasmon coupling

The last term in the Lagrangean $L$ represents the coupling between the longitudinal plasmons and the ripplons, with coupling strength given by

$$\gamma_\ell = Ne^2 \frac{(\ell + 1)}{\ell + \varepsilon(\ell + 1)}. \quad (22)$$

For electrons on a flat helium surface, such a coupling between ripplons and longitudinal plasmons/phonons was derived by Fisher et al. [4] and detected experimentally [3]. For electrons on the inner surface of a deformed bubble, we find a similar coupling, but only ripplon and longitudinal plasmon modes which have the same angular momentum couple to each other. After the diagonalisation of the ripplon-plasmon part of the Lagrangian $L$, we arrive at the eigenfrequencies,

$$\Omega_{1,2}(\ell) = \sqrt{\frac{1}{2} \left[ \omega_p^2(\ell) + \omega_r^2(\ell) \right]} \pm \sqrt{\frac{1}{2} \left[ \omega_p^2(\ell) - \omega_r^2(\ell) \right]^2 + 4\gamma_\ell^2}. \quad (23)$$

In Fig. 1, we show the eigenfrequencies $\Omega_1(\ell)$ and $\Omega_2(\ell)$ for $N = 10^5$ as a function of $\ell$ and $p$. The frequency $\Omega_2(\ell)$ is close to the ripplon frequency derived for an MEB within the approximation of a conducting surface [2]. Consequently, this frequency can be attributed to the ripplon modes, renormalized due to the ripplon-plasmon mixing. The admixture with the longitudinal plasmon mode is weak. The other branch of oscillations with the frequencies $\Omega_2(\ell)$ can be related to the longitudinal plasmon mode admixed with a small component
of ripplon nature. In typical multielectron bubbles, the difference in frequency between the ripplon and the plasmon modes weakens the coupling between these modes. Unlike for electrons on a flat surface [4] the mixing is weak. Finally, note that the multielectron bubble will be stable when both $\Omega_1^2$ and $\Omega_2^2$ are positive. This condition is equivalent to

$$\omega^2_{p}(\ell)\omega^2_{r}(\ell) > \gamma_{\ell}$$  \hspace{1cm} (24)

By substituting the results (19), (17) and (22) in the inequality (24) it can easily be seen that when the radius of the bubble is equal to the equilibrium radius, the ripplon-plasmon mixing does not change the criterion of stability for a MEB formulated in Ref. [2].

IV. CONCLUSIONS

In this communication we studied the longitudinal plasmon-ripplon modes in a multielectron bubble. The central result is the Lagrangean (14) with the ripplon and plasmon frequencies (17), (19) and the coupling strength (22). Unlike for electrons on a flat surface, we find that the mixing of the modes is weak for typical multielectron bubbles, because the bare longitudinal plasmon and ripplon frequencies are different ($\omega_p(\ell) \gg \omega_r(\ell)$). The conditions for bubble stability discussed in Ref. [2] remain unaltered by the ripplon-plasmon mixing. The analytic expressions for the longitudinal plasmon frequencies and the coupling strength as a function of the angular momentum allow us to derive a compact formula describing

![Fig. 1](image)

FIG. 1: Vibrational eigenfrequencies of an MEB in liquid helium, $\Omega_1 (l)$ (phonon modes) and $\Omega_2 (l)$ (renormalized ripplon modes) given by Eq. (23) for $N = 10^5$ as a function of $l$ and of $p$. Inset: a schematic picture of the directions of motion for phonons and ripplons in an MEB. The ripplons are excitations of the helium surface (with typical frequencies in the MHz-GHz range), while the phonons are related to the motion of electrons (with typical frequencies in the THz regime) tangential to the bubble surface. Note the different scales for $\Omega_1$ and $\Omega_2$. 

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the charge redistribution taking place on a deformed bubble. For a bubble deformation in a particular spherical harmonic mode \( \{ \ell, m \} \), the charge will redistribute itself according to the same spherical harmonic mode, and the amplitudes of deformation will be proportional to each other.

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