

Plasma Oscillations in Single Walled and Double Walled Fullerenes

¹Binumole P. Kuriakose, ²K. A. Vijayalakshmi

¹R&D Centre, Bharathiar University, Coimbatore, India

²Dept. of Physics, Sri Vasavi College Erode, India

Abstract: This paper presents a brief study of plasma oscillations of high and low frequency in single walled and multiwalled fullerenes. The electron and ion components of each shell of fullerenes are regarded as the two species plasma system. The equilibrium density of electrons and ions changes to perturbed densities due to the plasma oscillations. Quantum hydrodynamic model and classical expression respectively are used for the calculations for it. Quantum Ion acoustic Waves and plasmon oscillations occur in the system. It is seen that plasmon frequency of the system decreases as the radius of the fullerene system increases. The Quantum Ion Acoustic Wave frequency is dependent on the quantum effects which are the speed of propagation of the density disturbance and quantum pressure.

1. Introduction

Fullerenes are molecular carbon nanostructures.⁽¹⁾ They consist of a spherical, ellipsoid, or cylindrical arrangement of dozens of carbon atoms. Theoretical and experimental studies were done on the physical characters of fullerenes. The optical spectrum of the C₆₀ molecule was studied by Larsson et al.⁽²⁾ Bertsch et al.⁽³⁾ calculated the electromagnetic response function of C₆₀ cluster and discussed collective excitations. The excitations are found to occur at high energies of 20 eV. Barton and Eberlein⁽⁴⁾ used a two-fluid hydrodynamic model to study the plasmon spectrum in fullerenes C₆₀ and C₇₀. Michalewicz and Das⁽⁵⁾ considered C₆₀ molecule as a hollow sphere with finite thickness for studying the collective excitations. Long and Bose⁽⁶⁾ used the two dimensional model to calculate the dispersion relations for the collective oscillations.

Another collective resonance in C₆₀ was observed at almost 40 eV which was proved by Scully.⁽⁷⁾ Later Quantum Hydrodynamic Model was used to describe the electron Ion quantum Plasma oscillations in C₆₀ molecule by Moradi⁽⁸⁾. The C₆₀ molecule is very like hollow shell made from a single hexagonal layer from a graphite crystal, i.e., graphene. Thus, C₆₀ molecule support plasma oscillations, closely related to the plasma oscillations seen in graphene. Nevertheless, no explicit calculation can be found for electron-ion plasma oscillations in C₆₀ molecule. The multishell fullerenes are nano scale systems made from graphene sheets curved to form super lattices of spherical symmetries. Plasmon excitations of high energy electrons in multishell fullerenes are studied by Stockley et al.⁽⁹⁾ Yannouleas et al.⁽¹⁰⁾ studied the plasmon excitations of π and σ electrons in single and multishell fullerenes and it was found that the dispersion characteristics of the multishell fullerenes were found more complex.

2. Plasmon Oscillations in Fullerenes

A rigid inert background with dynamical properties is assumed for the dispersion studies of the fullerenes. The perturbed electron number density is deduced by Quantum Hydro Magnetics which can provide more clear dispersion relations. In a compressible background QIAW

oscillations are obtained. Graphene's lattice structures are neglected here so that plasmon excitations of sufficiently long wavelengths dominate the 2D electron fluid response.

A multishell fullerene consists of N concentric spheres of infinitesimally thin shell thickness and radii given by a_1, a_2, \dots, a_N . Each layer of the system is considered to be consists of electron fluid and ion fluid superimposed at $r = a_j$ where $1 \leq j \leq N$. The charges are considered to be e and Ze respectively for electrons and ions. Charge neutrality on each layer requires that the equilibrium density of electrons and ions are given by $n_i^0 = n_0$ and $n_e^0 = Zn_0$. Spherical coordinates are used to represent a point in space and the point in a particular surface is represented as $x_j = (a_j, \theta, \phi)$. The perturbed density of the homogeneous electron and ion fluids on the j^{th} layer are given by $n_{ej}(x_j, t)$ and $n_{ij}(x_j, t)$. The density perturbations occur due to the plasma oscillations with frequency ω .

The behaviors of the plasma under consideration are explained by equation of continuity and equation of momentum. The perturbed electron number density is deduced by QHM and ion density by classical expression

$$\frac{\partial u_{ej}(x_j, t)}{\partial t} = \frac{e}{m_e} \nabla_j \varphi(x, t) - \frac{\alpha}{Zn_0} \nabla_j n_{ej}(x_j, t) + \frac{\beta}{Zn_0} \nabla_j [\nabla_j^2 n_{ej}(x_j, t)] \quad (1)$$

$$\frac{\partial u_{ij}(x_j, t)}{\partial t} = \frac{Ze}{-m_i} \nabla_j \varphi(x, t) \quad (2)$$

$$\text{Here } \nabla_j = \frac{1}{a_j} \hat{e}_\theta \frac{\partial}{\partial \theta} + \frac{1}{a_j \sin \theta} \hat{e}_\phi \frac{\partial}{\partial \phi}$$

Which differentiates only tangentially to the surface. The first term on the right hand side of equation (1) is the force on electron fluid due to the tangential component of the electric field. The second and third term are the parts of the internal interaction force in the electron gas. The values of α and β are given by

$$\alpha = \frac{\pi Z n_0 \hbar^2}{m_e^2}$$

$$\beta = \frac{\hbar^2}{4m_e^2}$$

α is the square of the speed of propagation of density disturbances in a uniform 2D homogeneous Fermi electron fluid and β is the quantum pressure. Equation (1) and (2) refers to a self consistent potential due to the perturbations of the electron and ion fluid densities on all fullerene surfaces given by

$$\varphi(x,t) = e \sum_k \int d^2 x'_k \frac{zn_{ik}(x'_k, t) - n_{ek}(x'_k, t)}{|x-x'_k|} \quad (3)$$

Where $x'_k = (a_k, \theta', \varphi')$ and $d^2 x'_k = a_k^2 \sin \theta' d\theta' d\varphi'$

The Fourier Legendre transform $\tilde{A}(l,m,\omega)$ of an arbitrary function $A(\theta, \varphi, t)$ is given by

$$A(\theta, \varphi, t) = \sum_{l=0}^{\infty} \sum_{m=-l}^l \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \tilde{A}(l,m,\omega) Y_{lm}(\theta, \varphi) e^{-i\omega t} \quad (4)$$

The Coulomb potential in spherical co-ordinates is given by⁽¹¹⁾

$$\frac{1}{|x-x'|} = \sum_{l=0}^{\infty} \sum_{m=-l}^l g_l(r, r') Y_{lm}^*(\theta', \varphi') Y_{lm}(\theta, \varphi) \quad (5)$$

Where $g_l(r, r') = \frac{4\pi}{2l+1} \frac{r_{<}^l}{r_{>}^{l+1}} r_{<} = \min(r, r')$ and $r_{>} = \max(r, r')$

Using equations (4) and (5) in the Fourier Transforms of the perturbed electron and ion density are given by

$$\tilde{n}_{ej}(l,m,\omega) = \frac{zen_0 l_{aj}^2 \tilde{\varphi}(r,l,m,\omega)}{m_e(\omega^2 - l_{aj}^2 \alpha - l_{aj}^4 \beta)} \quad (6)$$

$$\tilde{n}_{ij}(l,m,\omega) = \frac{zen_0 l_{aj}^2 \varphi(r,l,m,\omega)}{m_i \omega^2} \quad (7)$$

Where $l_{aj}^2 = \frac{l(l+1)}{a_j^2}$ and

$$\tilde{\varphi}(r,l,m,\omega) = e \sum_j g_l(r, a_j) a_j^2 [Z \tilde{n}_{ij}(l, m, \omega) - \tilde{n}_{ej}(l, m, \omega)]$$

Here a rigid inert background with dynamical properties are assumed and the equations can be solved to obtain the fourierlegendre transforms of the individual induced electron densities \tilde{n}_{ej} .

$$[\omega^2 - (\alpha + l_{aj}^2 \beta) l_{aj}^2] \tilde{n}_{ej}(l,m,\omega) - \sum_k G_{jk}(l) \tilde{n}_{ek}(l, m, \omega) = 0 \quad (8)$$

Where $G_{jk}(l) = \frac{e^2 Z n_0}{m_e} l(l+1) \left(\frac{a_k}{a_j}\right)^2 g_l(a_j, a_k)$

j and k denotes different shells of the multi shelled structure.

The roots of equation(8) defines plasmon oscillations that are separated into a high frequency $\omega_+(l)$ and low frequency $\omega_-(l)$ for each l.

In the case of a single shell i.e N=1, the plasmon frequency is given by

$$\omega_j^2(l) = \alpha l_{aj}^2 + \beta l_{aj}^4 + \frac{zn_0 e^2 a_j^2}{m_e} l_{aj}^2 g_l(a_j, a_j) \quad (9)$$

The equation (9) shows that the plasmon frequency of a single shell depends on the sphere radius a_j and the variable l. It is clearly seen that as the radius increases the

plasmon frequency of the system decreases and tend to vanish as $a_j \rightarrow \infty$. The plasmon frequency becomes larger as the variable l increases.

When there are two spherical shells, there are two modes with different frequencies. It is given by the expression,

$$\omega_{\pm}^2(l) = \frac{\omega_1^2(l) + \omega_2^2(l)}{2} \pm \sqrt{\left(\frac{\omega_1^2(l) - \omega_2^2(l)}{2}\right)^2 + \left(\frac{e^2 Z n_0 a_1 a_2}{m_e}\right)^2 l_{a1}^2 l_{a2}^2 g_l^2(a_1, a_2)} \quad (10)$$

The last term in the square root gives the interaction between the two spherical shells.

3. Quantum Ion Acoustic Waves in Fullerenes

QIAW oscillations are coupled low frequency wave oscillations in multishell fullerenes. The small electron inertial force is taken into account i.e, $\frac{m_e}{m_i} \ll 1$. The LHS of (1) can be neglected under low frequency disturbance. The coupled low frequency equation will then become

$$\tilde{n}_j(l, m, \omega) - \sum_k G_{jk}(m, \omega) \tilde{n}_k(l, m, \omega) = 0 \quad (11)$$

Where $G_{jk}(l) = \frac{e^2 Z n_0}{m_e} a_k^2 g_l(a_l, a_k) \left[\frac{Z m_e l_{aj}^2}{m_i \omega^2} - \frac{1}{\alpha + \beta l_{aj}^2} \right]$

Equation (11) will give the N positive roots for ω defining the QIAW oscillations which are clearly separated into a high frequency $\omega_+(l)$ and a low frequency $\omega_-(l)$ for each l. For a single cell, N=1, the QIAW frequency is given by

$$\omega_j^2 = \frac{Z m_e}{m_i} (\alpha + \beta l_{aj}^2) l_{aj}^2 \left[1 + \frac{\alpha + \beta l_{aj}^2}{Z n_0 e^2 a_j^2} \frac{1}{m_e g_l(a_j, a_j)} \right]^{-1} \quad (12)$$

The equation shows that the QIAW frequency of a single shell the variable depends on l and the sphere radius a_j and it becomes smaller as the radius increases. The dimensionless QIAW dispersions can be graphically plotted with the variable l in the X axis and $\frac{\omega}{\omega_s}$ in the Y axis.

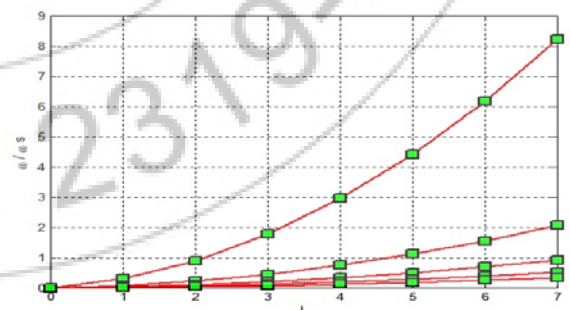


Figure 1: Dispersion relation for a value of l=1-8

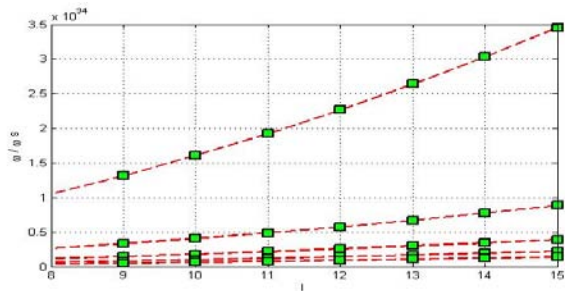


Figure 2: Dispersion relation for a single walled fullerene for a value of $l=8-15$

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[11] Jackson J D *Classical Electro dynamics* 1962 (new York Wiley)

If $N=2$ the case of two spherical shells comes into the position. Then the resulting 2×2 matrix on the left-hand side of the equation (11) gives the eigen value equation for the resonant frequencies of the QIAW oscillations in the coupled 2D layers, with the following dispersion relation.

$$\left[\left(\frac{Zm_e}{m_i} \right)^2 A - C \right] \omega^4 - \left[\left(\frac{Zm_e}{m_i} \right)^2 A (\omega_1^2(l) + \omega_2^2(l)) - B \left(\frac{Zm_e}{m_i} \right) C \right] \omega^2 + A \left(\frac{Zm_e}{m_i} \right)^2 [\omega_1^2(l)\omega_2^2(l) - C] = 0 \quad (13)$$

$$\begin{aligned} \text{Where } A &= (\alpha + \beta l_{a1}^2)(\alpha + \beta l_{a2}^2) l_{a1}^2 l_{a2}^2 \\ B &= \alpha(l_{a1}^2 + l_{a2}^2) + \beta(l_{a1}^4 + l_{a2}^4) \\ C &= \frac{\omega_1^2(l)\omega_2^2(l)g_l^2(a_1, a_2)}{g_l(a_1, a_1)g_l(a_2, a_2)} \end{aligned}$$

Where $\omega_j(l)$ are the frequencies of the individual QIAW oscillations on the spheres $j=1$ and 2 given by equation (12). When $a_1 = a_2$ the equation becomes that of single walled fullerene.

4. Conclusion

Here the dispersion relation for plasmon oscillations and QIAW frequencies for single walled and multi walled fullerenes are derived fluid theory and Poisson equations. The QIAW dispersion relations for single walled fullerenes are plotted against the variable l . The electrons are pulled along with ions and tend to shield out electric fields arising from the bunching of ions. The QIAW oscillations exist only when there are quantum effects. The graphs are in perfect explanation of the theoretical situation that as the radius increases the frequency decreases and tend to vanish at $a_j \rightarrow \infty$

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