C_{60} ELECTRON COLLECTIVE EXCITATION NATURE

V.V. Rotkin, R.A. Suris

Ioffe Physical–Technical Institute, 26, Politekhnicheskaya st., 194021 St.Petersburg,

Russia.

(March 15, 1996)

High frequency plasma excitation of C_{60}, which was detected by optical methods and EELS, is found to consist of two coupled components. A comparison with a graphite planar structure shows that electrons of σ and π valence bands with different symmetry participate in common oscillations. This oscillation energy is calculated within a simple model. We investigated some possible mechanisms of a strengthening of lower collective excitation frequency and found it to be similar to the acoustic plasma branch in the solids. The frequencies of two branches depend on the plasmon multipole index like the two-dimensional plasmon frequency depends on the wavevector.

I. INTRODUCTION

The problem of two-dimensional electron gas (2DEG) is in focus of attention during last decades. In particular, it was firmly stated that Coulomb coupling between two layers with 2DEG modifies the system response and leads to splitting

940

between two 2D-plasmon branches [1,2]. In this paper we consider in this paper a problem of coupling of 2D-plasmons on the surface of a conducting sphere. We suggest that the obtained result is applicable to the fullerene C_{60} molecule as well as to other spherical carbon nanoclusters. The approach considered here was successfully applied [3,4] to the recently discovered [5] bispherical C_{119} macromolecule and, probably, can be generalized to some quantum dot systems.

In relation to this problem we solved the model of Spherical Shell Quantum Well (SSQW) for C_{60} cluster [6–8] and obtained within the model that electrons, freely moving within a thin spherical shell, behave as a charged liquid at the frequency higher than all single electron transition frequencies. This system possesses some collective excitations [9,10] like a conducting sphere. In frame of SSQW model [7] we calculated plasmon frequency for quasi-spherical C_{60} molecule as the lowermost frequency of the dipole excitation of the total electron density of molecule. This frequency is triply degenerated. For the dimerized molecule with two SSQWs we considered a coupling between two spherical 2D-plasmons and obtained a new dipole excitation which frequency is splitted in the axial coupling field [3]. We showed that the model returns the well-known [1,2] result for two coupled metal planes in the contraction limit of the infinite radius of the spheres.

At the high frequency the spherical surface plasmon has to determine the optical response of the spherical nanocluster or the spectrum of electron energy losses. That is why it is interesting to clear how the simple hydrodynamic picture accords with the multi-component nature of σ and π electron gases in the fullerene.

It was suggested in [9] that two-component plasma on the sphere can be de-

941

scribed quantitatively with two phenomenological parameters: the restoring force for σ electron liquid and the displacement of the electron density from the mean radius of the sphere. However the last quantity was taken the same for both liquids that is crucial for Coulomb coupling between plasmons calculated in this paper. We point out that in the limit of "empty lattice" the restoring force should be zero, so the resulting frequencies of plasma oscillations in [9] remain to depend on the lattice potential strength, meaningless parameter while the continuous charged fluid approximation used.

It was analyzed that Coulomb interaction between two plasma liquids with the different symmetry gives nontrivial result. It will be shown below that considering within the free electron model both σ and π electrons without any additional restoring force one gets a new parameter with the dimensionality of frequency $e^2/\hbar C$, where e and $\hbar$ are the electron charge and Planck constant, and C is the capacitance of the system. It is not very useful for the planar 2DEG (or for an infinite graphite sheet) because of the inverse capacitance of the infinite plane capacitor decreases as its area. Though it is important for the spherical capacitor. Let us to evaluate this amount. When one takes all π electron shell density at the distance $a/2$ away from the mean radius, R, where the σ electron density is placed, the capacitance is $R(4R/a)$ with an accuracy of the small parameter $(a/2R)^2 \simeq 0.1$.

We stress that our model qualitatively represents the two-excitation character of plasma response of the fullerene as a result of the Coulomb mixing between plasmons, but it easily includes any additional refining terms. As a simple example the hydrostatic pressure will be considered which results in the dispersion of the

942

plasma frequency with the angular momentum of the excitation like the volume plasma frequency with the momentum of the plasmon.

The paper is organized as follows, useful results for SSQW plasmon will be given in the next section. The Sec.III deals with the Lagrangian formalism allowing us to include all interesting terms in the consideration. In the Sec.IV the model of C_{60} is considered. Last section contains the discussion of the model results.

II. PLASMON IN SSQW

Let us remember how the spherical plasmon was considered in frame of the spherical shell model of fullerene molecule [6–8]. In accordance with [8] we use the spherical oscillation of the electron density σ_{LM}. For the central symmetry of fullerene sphere we use the expansion of all quantities in the complete spherical harmonics [11] $P_{L}(r)Y_{L,M}(\Omega)$ those form a complete set on a sphere. The electric potential is the solution of Laplace equation for empty space inside, $\varphi^{\text{in}}(r < R)$, and outside, $\varphi^{\text{out}}(r > R)$, the spherical shell of molecule. We wrote it in multipole power expansion:

$$\varphi^{\text{in}}(r, \Omega) = \sum_{L,M} \frac{\varphi_{LM}}{R^L} |\mathbf{r}|^L Y_{L,M}(\Omega),$$

$$\varphi^{\text{out}}(r, \Omega) = \sum_{L,M} \varphi_{LM} R^{L+1} \frac{1}{|\mathbf{r}|}^{L+1} Y_{L,M}(\Omega).$$

At the spherical geometry a radial jump in electric field is given by:

$$\frac{2L+1}{R} \varphi_{LM} = 4\pi\sigma_{LM} \tag{1}$$

943

and we get response of the sphere in block-diagonal form [7] in subspace of angular momentum $L = \text{const}$. The next equation can be considered as the definition of χ, the response function:

$$\sigma_{LM} = -\chi_L \, \varphi^{act}_{LM} \, , \tag{2}$$

where $\varphi^{act}_{LM} = \{\varphi^{\text{xt}} + \varphi^{ind}\}_{LM} = \varphi^{\text{xt}}_{LM} + 4\pi R \sigma_{LM}/(2L+1)$ is the acting potential, L, M are the multipole power indexes (or the angular momentum and its projection onto z-axis). The selfconsintency of the calculation is provided by the including of the depolarization of the molecule through φ^{ind} in the full acting potential.

Like for 2DEG we get the eigenvalue equation for the single sphere excitation frequency arising from Eq.(1) and (2) under the zero external potential. Alternatively, each pole of the self-consistent polarizability, $\alpha_{LM}(\omega)$ is fixed by the frequency of the molecular plasma excitation with the angular momentum L. It is easily seen [7] from the explicit expression for the polarizability:

$$\alpha_L = -\frac{\sigma_{L,M}}{e\varphi^{\text{xt}}_{L,M}} = \frac{\chi_L}{1 + \frac{4\pi R}{2L+1}\chi_L} \, . \tag{3}$$

An explicit form of the spherical response function $\chi^L(\omega)$ could be obtained using the perturbation theory (see [7] for more details):

$$\chi_L(\omega) = \frac{e^2}{\hbar R^2} \sum_{n_i, \varsigma_i} \sum_{\Lambda, K} \frac{2f_\Lambda \, \omega_{K,\Lambda}}{\omega^2_{K,\Lambda} - \omega^2} \frac{(2\Lambda + 1)(2K + 1)}{4\pi} \begin{pmatrix} L & K & \Lambda \\ 0 & 0 & 0 \end{pmatrix}^2 \, , \tag{4}$$

where: $\hbar\omega_{K,\Lambda} = \frac{\hbar^2}{2mR^2}\left(K(K+1) - \Lambda(\Lambda+1)\right)$ is the energy of the single electron transition between the levels with the angular moments K and Λ within the free

944

electron (SSQW) model; m, n_i, ς_i are the electron mass, the radial quantum number and the spin of the electron in the ith spherical shell. The last multiplier is the Vigner symbol, it comes from the Coulomb-like matrix element integrated over the spherical angles with the spherical harmonics of the free electron wave functions [7].

We point out that the high frequency limit of the single molecule response function is fulfilled: $\dfrac{4\pi R}{2L+1}\chi_{\,L}(\omega) \simeq -\omega^2_{\,L}/\omega^2$. This expression is easily obtained from the classic charge liquid equations:

$$\begin{cases} m\dot{v} = -\nabla\varphi \\ \dot{\sigma} + n\nabla v = 0 \end{cases} \tag{5}$$

The first is a Newton low and the second is a linearized continuity equation. Assuming potential φ to be a harmonic function as before we get the response in multipolar expansion:

$$\sigma_{L,M} = \frac{nL(L+1)}{mR^2\omega^2}\varphi^{\text{act}}_{L,M} \ . \tag{6}$$

It is shown [12] that the response function and hence the polarizability of the spherical molecule with closed shell electron structure has no dependence on M-quantum number.

III. LAGRANGE APPROACH

A. Lagrangian of 2DEG in the plane

It is well known that plasma frequency of 2DEG can be obtained resolving the secondary quantizied Lagrange equation for the interacting electron and electric

945

fields. Here we reproduce this result in order to introduce some useful notations.

Classic Lagrange function for system of electrons with a density n_0 reads:

$$\int d^2 x \left(n \frac{mv^2}{2} - \frac{e\varphi\rho}{2} \right). \tag{7}$$

This function is easily quantized in terms of variables φ, the full electric potential, and ρ, fluctuating part of full density $n = n_0 + \rho$. They are connected by Gauss-Ostrogradskii theorem. It shows up as simple expression in Fourier transforms: $e\varphi_k = \frac{2\pi}{k}\rho_k$. The resulting quantum operator of Lagrangian is given by the next equation:

$$L = \frac{1}{2} \sum_k \left(nm \frac{\omega^2 \rho_k^2}{n^2 k^2} - \frac{2\pi}{k} \rho_k^2 \right) = \frac{1}{2} \sum_k \frac{m\omega_k^2}{nk^2} \left(\frac{\omega^2}{\omega_k^2} - 1 \right) \rho_k^2 \,, \tag{8}$$

where we subtract $\omega_k = \frac{2\pi n e^2}{m} k$, the frequency of 2DEG plasmon with a wavenumber k. The eigen-frequency equations are not coupled for the different k.

B. Lagrangian of spherical oscillations

First of all we reproduce our result for the sphere with a charged liquid on its surface from Sec.II. In comparison with the plane one changes the operator of the electron momentum in the plane with a ratio of the angular momentum to the curvature radius $\hat{L}/R$ and in the classic limit one obtains $k \to (l + 1/2)/R$ where $l + 1/2$ is the classic eigenvalue for the angular momentum operator. So we resolve the Lagrangian Eq.(7) in terms of spherical harmonics of surface charge density $\sigma_{l,m}$:

$$L_0 = \frac{1}{2} \sum_{l,m} \left(nm \frac{\omega^2 \sigma_{l,m}^2}{n^2 l(l+1)/R^2} - \frac{2\pi R}{l+1/2} \sigma_{l,m}^2 \right) = \frac{1}{2} \sum_{l,m} \frac{mR^2 \omega_l^2}{nl(l+1)} \left(\frac{\omega^2}{\omega_l^2} - 1 \right) \sigma_{l,m}^2 \,. \tag{9}$$

946

Here $\omega_l^2 = \dfrac{4\pi n}{mR}\dfrac{l(l+1)}{2l+1}$ is the frequency of the surface plasmon in SSQW ([4,7,8]). R is the radius of the infinitely thin SSQW. m and n are the mass and the average surface density of electrons in SSQW respectively. We note that the reducing of the expression $\dfrac{mR^2\omega_l^2}{nl(l+1)}$ yields to $\dfrac{4\pi R}{2l+1}$ that results from previous equation for ω_l.

To obtain the polarizability of the single molecule in the SSQW model we have to include the external field in the free Lagrangian:

$$L = L_0 + \frac{1}{2}\int e\varphi^{\text{xt}}\sigma = \frac{1}{2}\sum_{l,m}\frac{4\pi R}{2L+1}\left[\left(\frac{\omega^2}{\omega_l^2}-1\right)\sigma_{l,m}^2 - e\varphi_{l,m}^{\text{xt}}\sigma_{l,m}\right] \quad . \tag{10}$$

In this expression we surly omit an hermitian conjugated part as well as in all other expressions. We can prove that it is permissible and this simplifies the overview of the problem. We use notation σ^2 for the product $\sigma_{l,m}^\dagger\sigma_{l,m}$.

We stress that the kinetic energy of the electron liquid is proportional to its inverse response function according to Eqs.(6,9). Below we will substitute $1/\chi$ for the kinetic energy term in the Lagrangian.

Varying the Eq.(10) on the $\sigma_{l,m}^\dagger$ we get:

$$\frac{mR^2\omega^2}{nl(l+1)}\sigma_{l,m} - \frac{4\pi R}{2l+1}\sigma_{l,m} - e\varphi_{l,m}^{\text{xt}} = 0 \quad . \tag{11}$$

One should recognize in the first term the minus sigma divided by the response function from the Section II. Multiplying all terms by $-\chi$ we get:

$$\sigma_{l,m} + \chi_l\left(e\varphi_{l,m}^{ind} + e\varphi_{l,m}^{\text{xt}}\right) = 0 \quad , \tag{12}$$

where we connect $e\varphi^{ind}$ the induced potential from the sphere charge density fluctuation with its sigma through Gauss-Ostrogradskii low – Eq.(1). From the other

947

side we can divide the spherical component of the external potential by the minus proper component of the sigma that gives us the inverse spherical polarizability of the system:

$$\frac{1}{\alpha_l} = \frac{1}{\chi_l} + \frac{4\pi R}{2l+1} = \frac{4\pi R}{2l+1}\left(\frac{2l+1}{4\pi R\chi_l} + 1\right) \ . \tag{13}$$

C. Matrix Lagrangian

The Lagrangian in Eq.(10) can be considered as a matrix product of the Lagrangian matrix and the string and column vectors $(\sigma_{l,m})^T$ and $(\sigma_{l,m}, \varphi_{l,m}^{\text{xt}})$. The matrix Lagrangian has diagonal form, moreover it is block-identical matrix, $i.e.$ it is proportional to the unity matrix within each subspace of the fixed multipole power $l = $ const. We point out that the separation of matrixes in the different angular momentum subspaces is possible only at the lucky chosen the $SO(3)$ symmetry of the carbon cluster. More precisely that is really required is the "closed-shell" filling factor for all cluster single electron orbitals, at this assumption the electron system becomes invariant relatively to any rotation. We will discuss it at length elsewhere.

Let now the spherical oscillation of the electron density to have more than one component. This oscillation is conveniently considered as the multicomponent vector. Consequently, at l, m fixed, we can consider the Lagrangian matrix of 2×2 dimension multiplied by the two-component vectors of σ. Starting with the free Lagrangian such as in Eq.(9) one should write the potential energy in the form $\frac{4\pi R}{2l+1}(\sigma_1 + \sigma_2)^2$. Besides the direct self-acting terms $\sigma_{(i)}^\dagger \sigma_{(i)}$ the potential energy

948

has the crossed inter-acting terms $\sigma^{\dagger}_{(i)}\sigma_{(j)}$, where i, j denote σ or π electron liquid. The following Lagrangian

$$L_0 = \frac{1}{2}\sum_{i,j}\sum_{l,m}\left(\frac{-1}{\chi^i_l}\sigma^2_{l,m}(i)\ \delta_{ij} - \frac{4\pi R}{2l+1}\sigma^{\dagger}_{l,m}(i)\sigma_{l,m}(j)\right) \tag{14}$$

results in the matrix form, which reads:

$$\det\begin{vmatrix} -\dfrac{1}{\chi^{(1)}_l(x)} - \dfrac{4\pi R}{2l+1} & -\dfrac{4\pi R}{2l+1} \\[2ex] -\dfrac{4\pi R}{2l+1} & -\dfrac{1}{\chi^{(2)}_l(x)} - \dfrac{4\pi R}{2l+1} \end{vmatrix} = 0 \tag{15}$$

The problem is stated as the eigen-value problem for Lagrangian matrix respectively to the inverse response function argument $x_i = (\omega/\omega^{(i)}_l)^2$. Subtracting the common multiplier $\dfrac{4\pi R}{2l+1}$ outside the matrix one gets:

$$\begin{vmatrix} x_1 - 1 & -1 & \dfrac{2l+1}{4\pi R}e\,\varphi^{\text{xt}} \\[3ex] -1 & x_2 - 1 & \dfrac{2l+1}{4\pi R}e\,\varphi^{\text{xt}} \end{vmatrix} \tag{16}$$

Here we add the right part corresponding to the external field. The polarizabilities of the components of the surface density oscillations result from the resolving Eq.(16) with respect to φ^{xt}. The calculated polarizabilities are easily verified to be equal to the right part of the Eq.(3).

We point out that each pole of the α corresponds to each zero of the matrix determinant, it means that each eigen frequency of the multicomponent electron system gives a gain into the imaginary part of the selfconsistent polarizability of the system. The model has no additional excitation besides the spherical surface plasma excitation due to the special form of χ chosen (see Eq.(2)).

IV. TWO LIQUID MODEL

A. Why electrons in fullerene are distinct

The problem of quantum chemical hybridization of the electron orbital of a carbon in the nanocluster crystal lattice is not easy to solve. Each carbon has three nearest neighbors. The same situation in graphite results in $s - p^2$–hybridized electrons. Their bonds lie in the plane in a stick-and-ball model. When a carbon atom drops out from the initial plane position, it perforce gives a rise to an electron energy. In other words, it occurs the rehybridization in the curved graphite-like surface.

It is naturally to divide in two parts all valence electrons of carbon nanocluster similarly to the planar graphite. Three of four valence electrons of a carbon form 3 σ-bonds. They form the frame of the cluster, their electrons have low mobility. The rest one electron per carbon is highly mobile and participate in π-band building. The name σ or π is theoretical because of the rehybridization of bonds. The different kinetic properties of these two types of carriers allow us to distinguish between two charge liquids.

It was made for fullerene molecule in Ref. [9]. We will not address this result, but it worths to remind that authors of Ref. [9] took all two liquids on the sphere of a dumb-bell profile in a radial direction for the sake of simplicity. It is more natural to compute only π-electron liquid as the dumb-bell type. While a σ-electron density should have different symmetry relating to the tangent to sphere surface plane. Let us show that such clear assumption leads to significant change in the electron system

response.

The reduced free Lagrangian matrix (like in Eq.(16) without the right side) is obviously the sum of the diagonal matrix of the kinetic energy and the degenerate matrix of the potential energy. Consequently the first root of Eq.(15) is always zero. We emphasize that from mathematical point of view any changes in the first term of Lagrangian, in kinetic energy, can not alter the situation. Hence, in order to get physically correct result, when lower frequency is not zero, one does include a new potential energy part which differs for the two components (as it was done in Ref. [9] phenomenologically).

The origin of the strengthening of the lower plasmon should be revealed. We propose to account the difference in the radial integrals of the charge density overlap for the self- and inter-acting parts of the Coulomb energy. It will be shown in Sec.IV B that even the small overlap difference yields the essentially altered response. This quantity has the classical meaning mentioned in the introduction. It corresponds to the capacitance of the spherical capacitor with three-layers: the internal and external ones are connected.

B. Coulomb inter-action energy: Overlap function

In this section we will apply the Lagrange formalism developed in Sec.III B and III C to the two electron liquid model of the fullerene molecule.

The kinetic term in the Lagrangian reads:

951

$$\frac{1}{2} \sum_{i=\pi,\sigma} \sum_{l,m} -\frac{1}{\chi_l^{(i)}} \sigma_{l,m}^{(i)\,2} = \frac{1}{2} \sum_i \sum_{l,m} \frac{R^2 \omega^2}{l(l+1)} \frac{m_{(i)}}{n_{(i)}} \sigma_{l,m}^{(i)\,2} \tag{17}$$

where $1/\chi^{(\pi,\sigma)}$ is the inverse response of π (σ) electrons which is proportional to ω_l^{-2}, hence it is proportional to the electron effective mass to the density ratio $m_{(i)}/n_{(i)}$. Even if the electron mass in Eq.(17) is the same for two bands, the density of σ-electrons is thrice more than π:

$$L_0 = \frac{1}{2} \sum_{l,m} \left[\frac{\omega^2 m R^2}{n l(l+1)} \left(4\sigma_{l,m}^{(1)\,2} + \frac{4}{3}\sigma_{l,m}^{(2)\,2} \right) - \frac{4\pi R}{2l+1} \left(\sigma_{l,m}^{(1)} + \sigma_{l,m}^{(2)} \right)^2 \right], \tag{18}$$

here $n = n_\pi + n_\sigma$ - the total electron density. This expression leads to the Lagrangian matrix. On the diagonal there are the expressions which are identical to the secular equations for π (σ) modes: $-1/\chi_l(x) - \dfrac{4\pi R}{2l+1}$. One gets the simplest form for determined above eigenvalue problem for the dimensionless variable x by dividing all matrix on the characteristic Coulomb energy $\frac{4\pi R}{2l+1}$. We call this form of the Lagrangian matrix the reduced Lagrangian:

$$\begin{vmatrix} 4x - 1 & -1 \\ -1 & \frac{4}{3}x - 1 \end{vmatrix} \tag{19}$$

Without Coulomb coupling between two plasmon modes this equation returns free π (and σ) electron plasma frequency $- \sqrt{1/4}\,\omega_l$ (and $\sqrt{3/4}\,\omega_l$) where ω_l is the plasma frequency calculated for the total number of electrons (compare with the common multiplier in Eq.(18)). However, when considering plasma oscillation which includes the Coulomb force, one can not neglect the coupling terms. In the above presented consideration we believe that the electron liquid is two-dimensional or, equivalently, the electrons move only on the sphere surface. Actually, despite of the partial

952

rehybridization of the chemical bond in the fullerene, in the radial direction the σ-electron liquid is mainly concentrated in a vicinity of the mean radius of the cluster R. At the same time one naturally thinks that π-electron density has a node in the radial direction at R, because of the node of the atomic orbital. Hence these two charge densities are spatially separated. The separation distance $a/2$ is a parameter of the model and can be evaluated as the π-electron cloud width being about the carbon atom size ~ 1.5 Å.

The discussed Coulomb inter-action changes the non-diagonal reduced Lagrangian matrix elements from 1 to some quantity to be determined from the electrostatics of the two plasmon charges. We gave a classical estimation of the capacitance of the uniformly charged triple spherical capacitor in the introduction. Here we argue that considering the σ and π electrons on the spherical surfaces $r_\sigma = R$ and $r_\pi^\pm = R \pm a/2$, one gets the Coulomb integral between the modes in the following form:

$$
\frac{1}{R}\left(1 - \xi_l\right) = \int r_1^2 dr_1 \int r_2^2 dr_2 \sigma_1(r_1, l, m) \frac{r_<^l}{r_>^{l+1}} \sigma_2(r_2, l, m)
$$
$$
\simeq \tfrac{1}{R}\left(1 - l\tfrac{a}{4R} - (l+1)\tfrac{a}{4R} + \ldots\right)
\tag{20}
$$

here $\xi_l \simeq (2l+1)\dfrac{a}{4R}$ is the correction to the Coulomb inter-action to be included in Eq.(19) obtained at the small parameter $a/4R \ll 1$. It directly corresponds to the exponential multiplier in the Coulomb interaction between 2DEGs in a double-quantum-well structure (see [12] for more details).

Then the Lagrangian matrix reads as:

$$
\begin{vmatrix}
4x - 1 & -1 + \xi \\[2mm]
-1 + \xi & \frac{4}{3}x - 1
\end{vmatrix}
\tag{21}
$$

This matrix is readily solved relatively to the dimensionless frequency squared $x = \omega^2/\omega_l^2$. It has two roots with the finite frequency, how it has been clamed above. Thus, the correction to the Coulomb interaction due to the spatial separation of the electron liquids in the radial direction leads to the crucial changes in the plasma spectrum. We found two peaks without any additional terms included in the Lagrangian. However, in the next section we will discuss what additional feature can be considered within our approach.

C. Plasmons in σ and π bands

It is well known that the effective electron mass along the graphite plane is much smaller than the free electron mass: $m_\pi < m_o$. We assume m, the effective mass of the electron in σ-bands, is about m_o because of the essentially smaller dispersion in these bands. It is hard to calculate perfectly the effective mass ratio for π and σ electrons in the fullerene. We consider it as a phenomenological parameter in our model. Precisely speaking we take $m_\pi = m/\theta$. It changes the kinetic part in the Lagrangian like the above counted difference in the density of the σ and π electrons.

We estimated the mass ratio θ considering the experimental data for the dipole plasma frequencies for the C_{60}. It should be about 6, that seems not incredible when one compares with the graphite σ and π band structure.

Now, we will take into account that the electron liquid potential energy contains

954

the hydrostatic pressure term. The including of the term in the Lagrangian leads to the additional mixing of the plasmons. It will be considered in more details elsewhere [12] that this term in the potential energy results in the strengthening of the plasma frequency. When taking into account the pressure in the electron liquid, the second sound wave solution appears. It corresponds to the spatial dispersion term for the volume plasma frequency expression (see [1]). The 2D-plasmon frequency depends on the square root of its wavevector, in particular, it comes from the expression (8). While the surface plasma frequency ω_k disperses by itself, the "sound" term influences on the large wavevector asymptotic. The straightforward analogy with the 2D-plasmon gives us that the square root dependence of ω_l on the l/R becomes the acoustic linear dependence for the angular momentum much larger than the critical value.

This critical angular momentum is none other than the inverse effective Bohr radius $a_B^{-1} = m_{(i)} e^2 / \hbar^2$. Hence, the reduced Lagrangian matrix including the "capacitor" and "sound" terms reads as:

$$
\begin{vmatrix}
4x - 1 - \left(l + \dfrac{1}{2}\right) \dfrac{a_B^{(1)}}{R} & -1 + \xi - \left(l + \dfrac{1}{2}\right) \dfrac{a_B^{(1)}}{R} \left(\dfrac{\omega_l^{(2)}}{\omega_l^{(1)}}\right)^2 \\[4ex]
-1 + \xi - \left(l + \dfrac{1}{2}\right) \dfrac{a_B^{(2)}}{R} \left(\dfrac{\omega_l^{(1)}}{\omega_l^{(2)}}\right)^2 & 4x \left(\dfrac{\omega_l^{(1)}}{\omega_l^{(2)}}\right)^2 - 1 - \left(l + \dfrac{1}{2}\right) \dfrac{a_B^{(2)}}{R}
\end{vmatrix}
\tag{22}
$$

where $\left(\omega_l^{(1)} / \omega_l^{(2)}\right)^2 = n_1 m_2 / n_2 m_1 = \theta/3 \sim 2$.

V. RESULTS AND DISCUSSION

Thus we showed in Sec.IV B that the Coulomb coupling alters the frequencies of the coupled plasmons. To summarize we state that the new frequencies are found from the eigenvalues of the matrix (22). One can see that in order to eliminate the meaningless zero eigenvalue one should consider two correction terms ("capacitor" and "sound"). Depending on the parameters $m_1/m_2, a, n_2/n_1 = 3$ for σ and π electrons the solution has different form.

The formulated eigenvalue problem can be easy analyzed when neglecting one term considering the other. Let first the "sound" correction to be zero $a_B = 0$. Then the value of the lowermost $l = 1$ plasma frequency of π electrons is shifted up, while the σ plasmon is going down. It is seen from the dependence of the dimensionless root of the eigenvalue equation for the matrix (22) on the multipole index l. It is true until $l + 1/2 \ll 2R/a \sim 2 \div 3$. When increasing the $l > 3$ the dispersion of x on l changes sign. The σ plasmon gets more and more Coulomb correction and the dependence of ω_l on l becomes linear. The π electron plasma frequency goes to zero. It occurs at $l \sim 4R/a \sim 5$.

The including of the hydrostatic pressure correction leads to the smoothing of the dispersion of x on l. At definite values of the parameters it can be no solutions for the non-zero lower plasmon branch. It seems not to be the case of C_{60} cluster. The above discussed results of the model are schematically depicted in the figure.

956

---

[1] T. Ando, A.B. Fowler, F. Stern, Rev.Mod.Phys., **54**, N 2, 437 - 672 (1982).

[2] A.V.Chaplik, Zh.Eksp.Teor.Fiz., **60**, 1845 - 1852 (Sov. Phys. - JETP, **33**, 997 - 1000) (1971).

[3] V.V.Rotkin, R.A.Suris, "The coupled plasma oscillations on two conducting spheres joined", Proc.of International Symposium "Nanostructures: Physics and Technology-95", pp.210-213, St.Petersburg, Russia, 26-30 June 1995.

[4] V.V.Rotkin, R.A.Suris, "Multipole excitations of two joint conducting spheres: Application to C119 molecule", Sol.State Comm., v. 97, N 3, 183-186, 1995.

[5] G.B. Adams, J.B. Page, M. O'Keeffe, O.F. Sankey, Chem.Phys.Lett., **228**, 485-489; and cites [1,2] here (1994).

[6] V.V.Rotkin, R.A.Suris, Proceedings of IWFAC-93, p. 23, St.Petersburg, Russia,

957

4-9 October 1993; V.V.Rotkin, R.A.Suris, Proceedings of IWAMPT-94, p. 39, Chernogolovka, Russia, 13-17 June 1994.

[7] V.V.Rotkin, R.A.Suris, Fizika Tverdogo Tela (Sov. Phys. Sol. State), **36**, N 12, 1899 - 1905 (1994).

[8] V.V.Rotkin, R.A.Suris, Mol. Mat., **5**, 87-94 (1994).

[9] G. Barton, C. Eberlein, J. Chem.Phys. **95**, N 3, 1512-1517 (1991).

[10] M.T. Michalewicz, M.P. Das, Sol.St.Comm. **84**, N 12, 1121-1125 (1992).

[11] L. Robin, *Fonctions Spheriques de Legendre*, Gauthier–Villars, Paris, (1958), p.161.

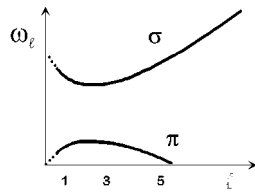[12] V.V.Rotkin, R.A.Suris, to be published.

FIG. 1. The schematical representation of the spectrum of the fullerene cluster plasma modes. The Coulomb coupling and the hydrostatic pressure in the electron gas are taken into account.

959