ELECTRON-ENERGY-LOSS SPECTROSCOPY OF THE C$_{60}$ MOLECULE

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In this work we present a theoretical study of EELS (electron-energy-loss spectroscopy) experiments on the C$_{60}$ molecule. Our treatment of the problem is based on the simple two-fluid model originally proposed for the description of plasma oscillations in graphite and fullerenes (see Refs. [1,2]). It is shown that in spite of the simplicity of the model the calculated intensities of the EELS peaks are in good agreement with experimental data which may indicate that the model can be used as a simple and effective tool for the investigation of the collective behaviour of electrons in fullerene systems.

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I. INTRODUCTION

Investigations of plasmons in small clusters of nanometer size range have a long history. Originally plasma oscillations were studied in small ($R = 2 \times 10$ nm) metal particles. It turned out that plasmons in metal clusters can be well described by linearized hydrodynamic equations[11]. The positions of plasmon peaks, their number and fast electron cross-sections calculated within these equations are in good agreement with experimental data. New possibilities of the investigation of plasmons in small objects appeared after the discovery of fullerenes and the cheap methods of their synthesis. Plasmon oscillations were observed in fullerene molecules[4,5], carbon nanometer size tubes[6] and multishell fullerenes[7].

An important feature of fullerenes is the fact that these molecules are more or less of a spherical form and their electrons can be considered to be confined to the surface of the sphere. Hence, it seems to be a good starting point to consider the C$_{60}$ molecule as a rolled up graphite plane. This approximation is expected to work well for the description of collective excitations.

The graphite plane consists of regular hexagons formed by carbon atoms. Each carbon atom is connected by 3 \( \sigma \)-bonds with its three neighbours, meanwhile \( \pi \)-electrons form a common \( \pi \)-system consisting of orbitals perpendicular to the plane. In linear response theory \( \pi \) and \( \sigma \)-electron subsystems can be considered to be independent with respect to the electrical field parallel to the graphite plane because dipole transitions from \( \pi \) to \( \sigma \)-orbitals are prohibited. This means that we can describe the plasmons in graphite whose wave vector is parallel to the plane as the plasmons in a “two-liquid” electron system; it is proposed that both the liquids move in a common average potential.

The graphite loss function \(-\text{Im}(1/\varepsilon_{\|})\) with respect to the external electric field perpendicular to the \( c \)-axis shows two peaks at 7 and 28 eV (see Refs.[12,13]). They are called \( \pi \)- and \( \sigma \)- (or “\( \pi + \sigma \)”) plasmons. Strictly speaking such a division is rather conventional. In fact, both \( \pi \)- and \( \sigma \)-electrons make a nonzero contribution to \( \sigma \)- and \( \pi \)-plasmons respectively.

In the articles[14,15] Cazaux used the two-fluid model in order to explain the results of EELS studies on graphite. The same idea was used in Ref.[16] for the description of plasmons in the C$_{60}$ molecule: it was proposed that \( \pi \)-electrons be affected only by induced electric potential, meanwhile \( \sigma \)-electrons be governed also by a restoring force. This force appears in the model via an effective description of a contribution of valent electrons to the polarizability[17,18].

Using the two-fluid model Barton and Eberlein predicted the existence of plasma oscillations in the C$_{60}$ molecule which can be classified by the spherical quantum numbers \( l \) and \( m \) and the index \( j \) \((j = 1, 2)\) which corresponds to \( \pi \)- and \( \sigma \)-plasmons[14]. However, the plasmons with large \( l \) cannot exist because of their strong damping.

EELS studies on C$_{60}$ (see Refs.[19,20]) show a peak at 6.5-7 eV (also called “\( \pi \)-plasmon”) and a rather broad “\( \sigma \)-plasmon” peak at 15-16 eV. The former peak is identified usually as the \( l = 1 \ \pi \)-plasmon, meanwhile the latter peak is considered to be a combination of the \( l = 1 \ \sigma \)-plasmon and probably the plasmons with \( l > 1 \). However the

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existence of multipole plasmons in C\textsubscript{60} is still not clear. In Ref\textsuperscript{[4]} a small peak at 28 eV was observed and identified as the \(l = 1\) \(\sigma\)-plasmon.

The main goal of this work is to calculate the cross-section of fast electrons scattered by a C\textsubscript{60} molecule and to show that the contribution of the plasmons with \(l = 2\) to the cross-section is enough in order to be observed in EELS experiments. In the model we use it is possible to separate contributions of different plasmons, i.e. one can obtain relative cross-sections. Using this approach we can not calculate plasmon damping. Widths of plasmon peaks will be taken from experimental data.

II. QUANTIZATION OF PLASMA OSCILLATIONS ON A SPHERE IN THE TWO-FLUID MODEL

We shall consider electrons in the C\textsubscript{60} molecule to be distributed over the surface of a uniformly charged sphere — the positive molecular core in the model. The Fourier transformation of the charge density \(\delta n(\omega)\) induced by the potential \(\Phi\) including both the external and induced potentials is given by the expression

\[
\delta n(\omega) = -\sum_{\alpha} e^2 n_m f_{\alpha} \left( \frac{\omega^2}{\omega^2 - \omega_\alpha^2} \Delta_{\parallel} \Phi(\omega) \right)
\]

where \(n\) is the surface \(\pi\)- and \(\sigma\)-electron density, \(f_{\alpha}\) and \(\omega_{\alpha}\) are respectively the oscillator strength and electron energy levels. The index \(\alpha\) numerates the electron states. \(\Delta_{\parallel}\) denotes the angular part of the Laplas operator: in the model we use electrons cannot move perpendicularly to the surface.

As the \(\pi\)-electron subsystem of graphite is semimetal, we can use the following approximation for the \(\pi\)-electron contribution to the induced charge density:

\[
\delta n_{\pi}(\omega) = e^2 N \frac{1}{4\pi m R^2} \omega^2 \Delta_{\parallel} \Phi(\omega).
\]

Here we used the sum rule \(\sum_{\alpha} f_{\alpha \pi} = 1/4\) for the oscillator strength of \(\pi\)-electrons in C\textsubscript{60}. \(N = 60\) and \(R = 0.4\) nm are the number of \(\pi\)-electrons and the effective radius of the sphere respectively.

On the other hand, the \(\sigma\)-electron subsystem of graphite is dielectric with the gap of order 12 eV (see Ref\textsuperscript{[11] \(\omega_\sigma\)). The oscillator strength of the \(\sigma\)-electron subsystem has a maximum at \(\omega_{\sigma} \approx 16\) eV. Hence we can use the following approximate expression for the \(\sigma\)-electron contribution

\[
\delta n_{\sigma}(\omega) = 3e^2 N \frac{1}{4\pi m R^2} \omega^2 \Delta_{\parallel} \Phi(\omega).
\]

Here we used the sum rule \(\sum_{\alpha} f_{\alpha \sigma} = 3/4\) for the \(\sigma\)-electron subsystem. \(\omega_{\sigma} = 16\) eV is the characteristic transition frequency of \(\sigma\)-electrons.

The \(\Phi\)-field is connected with the total induced charge density \(\delta \sigma = \delta n_{\pi} + \delta n_{\sigma}\) through the Gauss theorem

\[
\frac{\partial \Phi}{\partial r} \big|_{r=R^+} - \frac{\partial \Phi}{\partial r} \big|_{r=R^-} = 4\pi \delta \sigma.
\]

It is convenient to expand the density fields in Fourier series on the sphere

\[
\delta n_{\pi} = \sum_{lm} a_{lm} Y_{lm}(\phi, \theta), \quad \delta n_{\sigma} = \sum_{lm} b_{lm} Y_{lm}(\phi, \theta), \quad \delta \sigma = \sum_{lm} s_{lm} Y_{lm}(\phi, \theta).
\]

Then using the multipole expansion of the potential \(\Phi\)

\[
\Phi = \left\{ \begin{array}{ll}
\sum_{lm} \phi_{lm} (r/R)^l Y_{lm}(\phi, \theta) & : r < R, \\
\sum_{lm} \phi_{lm} (R/r)^{l+1} Y_{lm}(\phi, \theta) & : r > R
\end{array} \right.
\]

we can rewrite (\ref{5}) in the following form

\[
\phi_{lm} = -\frac{4\pi R}{2l+1} s_{lm} = \frac{4\pi R}{2l+1} (a_{lm} + b_{lm})
\]

which gives for (\ref{2}) and (\ref{3}) (see also Refs.\textsuperscript{[11] \(\delta\))}
\[ \dot{a}_{lm} = -\Omega_l^2 (a_{lm} + 3b_{lm}) \] (7)

\[ \dot{b}_{lm} = -3\Omega_l^2 (a_{lm} + b_{lm}) - \omega_\sigma^2 b_{lm} \] (8)

where

\[ \Omega_l^2 = \frac{4\pi^2 \sigma_0 l(l+1)}{mR^2}, \quad \sigma_0 = \frac{N}{4\pi R^2}. \] (9)

It can be easily verified by straightforward calculation that equations (7) and (8) are the equations of motion corresponding to the Hamiltonian

\[ H = \frac{m\sigma_0}{2} \sum_{lm} \dot{A}_{lm} \dot{A}_{l,m} + \frac{3m\sigma_0}{2} \sum_{lm} \dot{B}_{lm} \dot{B}_{l,-m} + \]

\[ \frac{m\sigma_0}{2} \sum_{lm} \Omega_l^2 A_{lm} \dot{A}_{l,m} + \frac{3m\sigma_0}{2} \sum_{lm} (3\Omega_l^2 + \omega_\sigma^2) \dot{B}_{lm} \dot{B}_{l,-m} + \]

\[ \frac{m\sigma_0}{2} \sum_{lm} 3\Omega_l^2 (A_{lm} B_{l,-m} + A_{l,-m} B_{lm}) \] (10)

where the amplitudes \( A_{lm} \) and \( B_{lm} \) are the \((l,m)\)-components of the displacement fields \( r_1 \) and \( r_2 \) of the electron liquids which are connected with \( a_{lm} \) and \( b_{lm} \) by the following equations

\[ a_{lm} = \frac{\sigma_0}{4} \sqrt{\frac{l(l+1)}{R^2}} A_{lm}, \quad b_{lm} = \frac{3\sigma_0}{4} \sqrt{\frac{l(l+1)}{R^2}} B_{lm}. \] (11)

The coefficients in equation (10) are chosen so as to keep it equal to the hydrodynamic Hamiltonian

\[ H = \frac{m\sigma_0}{2} \int \dot{r}_1^2 dS + \frac{3m\sigma_0}{2} \int \dot{r}_2^2 dS + \frac{3m\sigma_0 \omega_\sigma^2}{2} \int r_2^2 dS + \frac{1}{2} \int \phi \delta \sigma dS \] (12)

Hamiltonian (10) is nondiagonal in the basis consisting of the functions \( Y_{lm}(\phi, \theta) \) because of the last term. On the other hand, this term does not mix amplitudes with different \( l \) and \( |m| \) because of the spherical simmetry of the model, hence the above Hamiltonian can be diagonalized by the diagonalization of each “block” with a number \( lm \).

We have shown that the problem can be reduced to the diagonalization of the Hamiltonian

\[ \frac{m\sigma_0}{2} \dot{A}^2 + \frac{m\sigma_0}{2} \dot{B}^2 + \frac{m\sigma_0}{2} \Omega_l^2 A^2 + \frac{3m\sigma_0}{2} (3\Omega_l^2 + \omega_\sigma^2) B^2 + 3m\sigma_0 \Omega_l^2 AB \] (13)

which can be done by the following transformation

\[ \sqrt{m\sigma_0} A = Q_1 \cos \phi_l - Q_2 \sin \phi_l \] (14)

\[ \sqrt{3m\sigma_0} B = Q_1 \sin \phi_l + Q_2 \cos \phi_l \] (15)

where the angle \( \phi_l \) can be found from the condition of the disappearance of the nondiagonal term in Hamiltonian (13) expressed in terms of the new coordinates \( Q_1 \) and \( Q_2 \). The substitution of (14) and (15) into equation (13) gives

\[ \cot 2\phi_l = -\frac{2\Omega_l^2 + \omega_\sigma^2}{2\sqrt{3}\Omega_l^2}. \] (16)

The eigenfrequencies \( \Omega_{l,j} \) are given by the following equation

\[ (\Omega_l^2)_{1,2} = \frac{1}{2} \left( 4\Omega_l^2 + \omega_\sigma^2 \pm \sqrt{4\Omega_l^2 \omega_\sigma^2 + \omega_\sigma^4 + 16\Omega_l^4} \right). \] (17)

One can clearly see that charge oscillations can be classified by the numbers \( l \) and \( m \) and the index \( j \) corresponding to the lower and upper roots of equation (17). The fact that the eigenfrequencies do not depend on the number \( m \) is due to the spherical symmetry of the considered model.
Using (11) we can find quantum mechanical operators for amplitudes $A_{lm}$ and $B_{lm}$. Substituting them into the equation for classical charge density fluctuations we obtain the operator corresponding to this quantity

$$
\delta \hat{\sigma}(\phi, \theta) = -\frac{e}{R^2} \sqrt{\frac{\hbar \sigma_0}{2m}} \sum_{lm} \sqrt{l(l+1)} Y_{lm}(\phi, \theta) \left( \frac{g_1(\phi_l)}{\sqrt{\Omega_{l,1}}} (\delta_{l,1} + \delta_{l,-1}) + \frac{g_2(\phi_l)}{\sqrt{\Omega_{l,2}}} (\delta_{l,2} + \delta_{l,-2}) \right)
$$

(18)

where $\delta_{lm,j}$ is the annihilation operator of the plasmon with numbers $lm, j$. Hence we can use the Born approximation. The quantitative criterium of the applicability of the Born approximation will be discussed further. The angle $\phi_l$ is chosen to be the minimal positive root of equation (16).

III. FAST ELECTRON SCATTERING

In this section we will obtain the expression for the scattering amplitude of fast electrons due to charge density fluctuations in $C_{00}$. In the secondary quantization representation the electron-plasmon interaction can be written as follows

$$
\hat{H}_{int} = \int \hat{\Psi}^\dagger (-e \hat{\Phi}) \hat{\Psi} dr.
$$

(20)

Here $\Psi = \sum_k e^{ikr} a_k/\sqrt{V}$ is the electron field operator, $\Phi$ is the potential corresponding to the electron surface density distribution $\delta \hat{\sigma}(\phi, \theta)$, i.e. $\Phi$ is the solution of the Poisson equation with the charge density $\delta(r-R)\delta \hat{\sigma}(\phi, \theta)$. Hence $\Phi$ and $\delta \hat{\sigma}$ are connected by the following equation

$$
\Phi = \int \frac{\delta(r' - R)\delta \hat{\sigma}(\phi', \theta')}{|r - r'|} dr'.
$$

(21)

Substituting equation (21) and the explicit expressions for the electron field and the electron density fluctuation operators (see (18)) into equation (20) we obtain the expression for the electron-plasmon interaction

$$
\hat{H}_{int} = -\sum_{kk'} \left[ \frac{4\pi^2 e^2}{|k - k'|^2} \sqrt{\frac{\hbar \sigma_0}{2m}} \right] \sum_{lm,j} \sqrt{l(l+1) \Omega_{l,j}} g_j(\phi_l) j_i \left( |k - k'| R \right) Y_{lm} \left( \frac{k - k}{|k - k'|} \right) (\delta_{l,1} + \delta_{l,-1}).
$$

(22)

We shall consider only the case of fast scattering electrons, i.e. the only relevant processes are those in connection with the creation of one plasmon. Hence we can use the Born approximation. The quantitative criterium of the applicability of the Born approximation will be discussed further.

In the initial state all the plasmon occupation numbers are equal to zero. Let us suppose that after scattering only the occupation number of the state $lm, j$ is equal to one. Defining the initial and final momenta of the scattering electron $k$ and $k'$ respectively we obtain the following expression for the modulus squared of the matrix element of $H_{int}$

$$
|\langle k; 0| \hat{H}_{int} |k'; lm, j \rangle|^2 = \left( \frac{4\pi^2 e^2}{2m} \sqrt{\frac{\hbar \sigma_0}{2m}} \right) \sum_{lm,j} \sqrt{l(l+1) \Omega_{l,j}} g_j(\phi_l) j_i \left( |k - k'| R \right) Y_{lm} \left( \frac{q}{q} \right) Y_{lm}^* \left( \frac{q}{q} \right)
$$

(23)

where $q = k' - k$.

As each eigenfrequency $\Omega_{l,j}$ is $2l + 1$-fold degenerated, we shall calculate the total cross-section for the transferred energy $\hbar \Omega_{l,j}$. Summing up the contributions of plasmons with given $l$ and $j$ over $m$ we get

$$
\frac{d\sigma_{q_j}}{d\Omega_{k'}} = \int \left( \sum_m |\langle k; 0| \hat{H}_{int} |k'; lm, j \rangle|^2 \right) \delta(\omega_{k'} + \Omega_{l,j} - \omega_k) \frac{2\pi V}{h^2} \frac{k'^2 dk'}{(2\pi)^3}.
$$

(24)
In the above equation $\omega_k = \hbar k^2/2m$, $v$ is the initial velocity of the electron.

Using the equation
\[ \sum_{lm} Y_{lm}(q/q) Y_{lm}^*(q/q) = (2l + 1)/4\pi \]  \hspace{1cm} (25)

(see Ref. 22) we get after some algebra
\[ \frac{d\sigma_{1,j}}{d\Omega_{k'}} = 8\pi \frac{e^4 e_0^2}{\hbar^3 v} (l+1)(2l + 1) \frac{g_2^2(\phi_l) \sqrt{k^2 - \frac{2m\Omega_{l,j}}{\hbar} j^2(l)[k' - k]/R}}{[k' - k]^4} \]  \hspace{1cm} (26)

IV. RESULTS AND DISCUSSION

In experiments one can measure the number of electrons reaching a detector, i.e. one measures the function (24) integrated over a certain solid angle. Let this solid angle be restricted by two cones of angles $\theta_1$ and $\theta_2$ respectively. In this case we can write for the total cross-section $\sigma_{1,j}(\theta_1, \theta_2)$
\[ \sigma_{1,j}(\theta_1, \theta_2) = \frac{2\pi e^4 N}{\hbar \Omega_{l,j}} (l+1)(2l + 1) g_3^2(\phi_l) \int_{|k-k'|R} \frac{j_l^2(x)}{x^{2l-3}} dx \]  \hspace{1cm} (27)

where $E = mv^2/2$, $|k - k'| = \sqrt{k^2 + k'^2 - 2kk' \cos \theta_i}$, $i = 1, 2$.

We shall consider the case of small transferred energy, i.e. $\omega_{k'} - \omega_k \ll \omega_k$. In principle, several different situations are possible. If both the angles $\theta_1$ and $\theta_2$ are small, the integration limits in the integral in (27) are also small and we can expand the spherical Bessel function into a Taylor series and take only the first term of the expansion. As $j_l(x) \approx x^l/(2l+1)!$ if $x \rightarrow 0$, we get
\[ \sigma_{1,j}(\theta_1, \theta_2) \sim \int_{|k-k'|R} x^{2l-3} dx \]  \hspace{1cm} (28)

and one can clearly see that the contribution of the plasmons with $l = 1$ is dominant. In the opposite situation $|k-k'| R \gg 1$, $i = 1, 2$ we can use the asymptotic expression for the spherical Bessel function $j_l(x) \approx \sin(x - \pi l/2)/x$ if $x \rightarrow \infty$. As the integration limits do not depend on $l$ significantly, we can see that the values of the integral in (27) are of the same order for different $l$. Hence the relative contributions of plasmons with different $l$ depend only on the factor $g_3^2(\phi_l)/\Omega_{l,j}$. In the case $|k-k'| R \sim 1$, $i = 1, 2$ it is possible to estimate the integral in (27) by the substitution $j_l(x) \sim x^l/(2l+1)!$. One can clearly see that only the contribution of the plasmons with small $l$ is relevant because of the rapidly increasing multiplier $(2(2l+1)!)$ in the denominator.

Strictly speaking the above analysis is not applicable for the plasmons with $l > 3$. The C$_{60}$ symmetry group is icosahedral and it does not have irreducible representations with a dimension of more than five. Hence the plasmons with $l > 3$ in the spherical model split and the classification of plasmons in the real C$_{60}$ molecule is different. We will not discuss further this difference because in the experiments which we will analyse only the contribution of the plasmons $l = 1, 2$ is relevant. On the other hand, the small size of C$_{60}$ leads to the strong damping of plasmons. The estimation for the spherical model in Ref. 23 shows that the plasmons with $l > 3$ could hardly exist.

We shall compare our results to the experiment from Ref. 24. In this work the number of electrons with $E = 1$ keV scattering in the solid angle $0.5^\circ < \theta < 2.5^\circ$ (i.e. $\theta_1 = 0.5^\circ$, $\theta_2 = 2.5^\circ$) per unit time was measured. The average cross-section
\[ \bar{\sigma}_{1,j}(\theta_1, \theta_2) = \sigma_{1,j}(\theta_1, \theta_2)/2\pi(\cos \theta_1 - \cos \theta_2) \]  \hspace{1cm} (29)

obtained using the spherical model for the plasmons with $l = 1 \ldots 5$ is shown in Table I. We used the value $\omega_{\sigma} = 16$ eV for the characteristic $\sigma$-electron transition frequencies and $R = 0.4$ nm for the radius of the sphere in the model. For the conditions of the analysed experiments $|k-k'| R \sim 1$, $i = 1, 2$ and one can clearly see that the contribution of the plasmons with $l > 2$ decays rapidly. This is in good agreement with the qualitative consideration. The $l = 1$ $\pi$- and $\sigma$-plasmons and the quadrupole $\sigma$-plasmon make the most significant contribution to the cross-section.
Plasmons in $C_{60}$ are characterized by strong damping due to the small size of the molecule: in small objects the energy of plasma oscillations per one electron $\hbar\Omega/N$ is strongly enhanced and leads to strong electron-electron scattering, meanwhile for bulk plasmons this energy is negligible. As energies of $\pi$- and $\sigma$-plasmons do not depend on $l$ significantly (see Table I), the parameter $\hbar\Omega/N$ is approximately the same for $\pi$- and $\sigma$-plasmons respectively and we can expect that the damping of plasmons does not depend on $l$ strongly.

In the above calculation of the cross-section we did not take into account plasmon damping. The characteristic inverse life time $\eta_l$ taken from the experiment is equal to 1.5 and 8 eV for $\pi$- and $\sigma$-plasmons respectively. The function

$$\sigma(\omega) = \sum_{l,j} \frac{1}{\pi} \frac{\delta_{l,j}(\theta_1, \theta_2)\eta_j}{(\omega - \Omega_{l,j})^2 + \eta_j^2}$$

(30)

is plotted in Fig. 1 together with the experimental curve. One can see that our results are in good agreement with the experiment.

As it can be seen from Table I and Fig. 1 the first peak on the graph corresponds to the dipole $\pi$-plasmon; the second peak corresponds to the dipole and quadrupole $\sigma$-plasmons. The contributions of the other plasmons are small. It has already been discussed that ratios $\sigma_{l,1}/\sigma_{l,2}$ are proportional in our case to ratios $(g_{l,1}^2/\Omega_{l,1})/(g_{l,2}^2/\Omega_{l,2})$. Note that although $\Omega_{l,1} < \Omega_{l,2}$ we have for the cross-sections $\sigma_{l,1} > \sigma_{l,2}$ and the ratio $\sigma_{l,1}/\sigma_{l,2}$ decreases if $l$ increases. This is the consequence of the fact that $g_{l,1} \to 0$ if $l \to \infty$; for large $l$ we can write using equation (16) $\cot \phi_l \to -1/\sqrt{3}$, hence $g_{l,1} \to 0$ (see (19)) and we obtain an interesting result: the ratio $\sigma_{l,1}/\sigma_{l,2}$ tends to zero if $l \to \infty$.

Let us discuss the condition of the applicability of the Born approximation. The Born approximation is valid if $|U|a \ll \hbar v$ (see Ref. 2) where $|U|$ and $a$ are the characteristic value of the potential and its radius respectively, $v$ is the velocity of the scattering particle. In our case the characteristic value of charge density fluctuations $\delta \bar{\sigma}$ is $(e/a^2)\sqrt{\hbar \sigma_0/m \Omega}$ where $\Omega \sim 10$ eV is the characteristic oscillation frequency. Hence $|U| \sim (e^2/a)\sqrt{\hbar \sigma_0/m \Omega}$ and the condition of the applicability can be written as follows

$$e^2 \sqrt{\hbar \sigma_0/m \Omega} \ll \hbar v.$$  

(31)

Substituting $\sigma_0 = N/4\pi R^2$, $a \sim R$, $R = 0.4$ nm into (21) we see that the Born approximation is applicable if the velocity of the scattering electron satisfies the condition $v \gg 2 \cdot 10^8$ cm/s. In the analysed experiments the electron velocity is $1.88 \cdot 10^9$ cm/s, i.e. the Born approximation is applicable.

Briefly summarizing we have shown that the simple two-fluid model used by the authors of Ref. 14 for the predictions of the collective spectrum of $C_{60}$ turned out to be good for the quantitative calculations of the intensities of the plasmon peaks in EELS spectra. The advantage of the description based on this model is that it is physically and technically simple and the model itself does not include any fitting parameters (with the exception of the decay rates which however cannot be properly calculated in more sophisticated techniques). All this makes the model useful for the quantitative consideration of the collective behaviour of electrons in more complicated fullerene systems.

| Type of plasmon | Plasmon frequency, eV | Cross section, nm$^2$/sr |
|-----------------|------------------------|--------------------------|
| $l=1, j=1$      | 5.95                   | 3.82                     |
| $l=1, j=2$      | 22.21                  | 9.56                     |
| $l=2, j=1$      | 6.68                   | 0.69                     |
| $l=2, j=2$      | 26.49                  | 5.16                     |
| $l=3, j=1$      | 7.03                   | 0.09                     |
| $l=3, j=2$      | 30.12                  | 1.35                     |
| $l=4, j=1$      | 7.23                   | 8.4 $\cdot 10^{-3}$     |
| $l=4, j=2$      | 33.35                  | 0.22                     |
| $l=5, j=1$      | 7.36                   | 5.6 $\cdot 10^{-4}$     |
| $l=5, j=2$      | 36.29                  | 0.024                    |
Fig. 1 Measured in Ref. 5 and calculated using equation (27) (solid line) inelastic cross-section with incident 1 keV electrons at 1.5° ± 1°.

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