On the interactions of the high energy photoelectrons with the fullerene shell

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The probability that photoionization of the caged atom in an endohedral system is accompanied by excitation of the fullerene shell is shown to be close to unity in broad intervals of the photoelectron energies. The result is obtained by summation of the perturbative series for the interaction between the photoelectron and the fullerene shell. This result means that the interaction between the photoelectron ejected from the cage atom and the fullerene shell cannot be described by a static potential, since inelastic processes become decisively important.

I. INTRODUCTION

Interaction of the high energy photons with endohedral atoms attracts much attention nowadays [1]-[4]. The inelastic processes in the fullerene shell (FS) caused by the electrons created in photoionization of the caged atom is one of the subjects of these studies. The calculations require understanding of the mechanism of interaction between the photoelectron and the electrons of the FS. In the present Letter we find the sum of the cross section of inelastic processes in the FS focusing on the energy behavior of the ratio

\[ r(E) = \frac{\sigma_A(E)}{\sigma_\gamma(E)}. \] (1)

Here \( \sigma_\gamma \) and \( \sigma_A \) are the cross sections of photoionization of the isolated atom, and that of the caged atom followed by inelastic processes in the FS (absorption cross section); the photoelectron carries away the energy \( E \). We demonstrate that in the broad intervals of the values of \( E \) the ratio \( r(E) \) is close to unity. In other words, the probability that photoionization of the caged atom is followed by an inelastic process in the FS is close to unity.
This result has consequences for the related problem of studying the wave function of the outgoing electron in photoionization of the caged atom. We can not consider the FS as just a source of an external field. Inelastic processes in the FS are important.

From the first sight the result looks surprising since the two main mechanisms of excitation and ionization of the FS are connected with small parameters. In the shake-off (SO) the electronic shell is moved to an excited state by the sudden change of the effective field caused by the incoming photon. Since FS is far from the caged atom, i.e. the FS radius \( R \) is big
\[
R \gg 1, \tag{2}
\]
the SO effects are of the order \( 1/R^2 \ll 1 \). (We employ the atomic system of units with \( e = m = h = 1 \)). The final state interaction (FSI) between the photoelectron and the bound ones is determined by its Sommerfeld parameter \( \xi = 1/v \) with \( v \) being the relative velocity of the photoelectron and the bound electrons. At high energies we can put
\[
\xi^2 \approx 1/2E \ll 1. \tag{3}
\]

Note however that while in FSI the electronic shell reacts upon creation of the hole as a whole, each bound electron is affected by FSI separately. Hence, the FSI parameter is rather \( \xi^2N \) with \( N \) being the number of the bound electrons, while in SO the electronic shell reacts upon creation of the hole as a whole and the SO effects do not depend on \( N \) directly. Thus, for light atoms \( \xi^2N \ll 1 \) already at the energies higher than several hundred eV. However, in the endohedral atoms, which are fullerenes stuffed with an atom inside, the number of electrons \( N \) is much larger and the FSI become weak at much larger energies. For example, there are \( N = 360 \) electrons in the fullerene \( C_{60} \), and \( \xi^2N \ll 1 \) only for \( E \gg 5 \) keV.

Fortunately, for the endohedral atoms one can sum the FSI power series in \( \xi^2N \) without assuming this parameter to be small. This is due to the large size of these systems - Eq.\(^2\). Localization of FS electrons in a thin layer
\[
R \leq r \leq R + \Delta, \tag{4}
\]
enabled us to calculate the sum of power series in \( \xi^2N \) in a model-independent way.

If the photoelectron energy \( E \) and thus its momentum \( p \) are large enough so that Eq.\(^3\) holds, the first step of the process is the photoionization of the caged atom, which takes place at the distances of the order \( r \sim 1/p \ll 1 \). After that the photoelectron passes the distances
of the order \( r \sim R \gg 1 \), interacting with the electrons of the FS. Thus, the amplitude \( F_x \) of the process with the final state of the electronic shell \( x \) contains the photoionization amplitude \( F_\gamma(E) \) as a factor, i.e. \[ F_x(E) = F_\gamma(E)T_x \] (5) with \( T_x \) being the amplitude of transition of electrons that belong to the FS. The accuracy of this equality is of the order \( V/E \) with \( V \) being the potential energy of the photoelectron in the field of the FS. In the lowest order of expansion in powers of \( 1/R \) we can neglect all the SO effects and thus \( T_x \) in Eq.(5) is in fact the FSI amplitude.

II. LOWEST ORDER TERMS

It is instructive to start with analysis of the lowest order terms. For the lowest order FSI amplitude of photoionization of caged atom followed by transition of the FS electrons from the initial state \( |\Psi_0\rangle \) to an excited state \( |\Phi_x\rangle \) is given by \[ T_x^{(1)} = \langle \Phi_x|U_1|\Psi_0\rangle, \] (6) where \( U_1 = \sum_k U_1(r^{(k)}) \), with \( k \) labeling the FS electron, \( U_1(r^{(k)}) \) is its interaction with the photoelectron in the lowest order of the FSI. One can write

\[
U_1(r^{(k)}) = \frac{1}{c} \int \frac{d^3f}{(2\pi)^3} G(f)g(f)e^{i(f|r^{(k)})},
\] (7)

where \( c \) is the speed of light and

\[
G(f) = \frac{-2}{2pf - i\nu}
\] (8)

is the free electron propagator, in which only the term proportional to the large momentum \( p \) is kept in denominator, \( g(f) = 4\pi/(f^2 + \lambda^2) \), \( \lambda \to 0 \). This provides

\[ U_1 = i\xi\Lambda; \quad \Lambda = \sum_k \ln(r^{(k)} - r^{(k)}), \] (9)

We shall see that the terms containing parameter \( \lambda \) form the Coulomb phase of the \( e - e \) scattering and will cancel in the final step.

The second order amplitude is \( T_x^{(2)} = \langle \Phi_x|U_2|\Psi_0\rangle \), where

\[
U_2 = \frac{1}{c^2} \sum_{k_1k_2} \int \frac{d^3f_{1}}{(2\pi)^3} \frac{d^3f_{2}}{(2\pi)^3} G(f_{1})g(f_{1})G(f_{1} + f_{2})g(f_{2})e^{i(f_{1}r^{(k_{1})})}e^{i(f_{2}r^{(k_{2})})}.
\] (10)

Using Eq.(8) for the Green function \( G \) and putting in the integrand \( G(f_{1})G(f_{1} + f_{2}) = (G(f_{1})G(f_{1} + f_{2}) + G(f_{2})G(f_{1} + f_{2})/2 = G(f_{1})G(f_{2})/2 \), we find that \( U_2 = U_1^2/2 \).
III. SUM OF THE POWER SERIES

One can see that this expression can be generalized for the case of arbitrary number \( n \) of interactions between the photoelectron and the FS. Introducing \( a_n = p(f_1 + f_2 + \ldots + f_n) \) we can write

\[
\frac{1}{a_1} \cdot \frac{1}{a_2} \cdots \frac{1}{a_n} = \frac{1}{n! a_1^n}. \tag{11}
\]

This equation, which can be proved by the induction method was used earlier for calculation of the radiative corrections in electromagnetic interactions \[8\]. Thus, the amplitude

\[
F_x = F_\gamma \langle \Phi_x | e^{i\xi \Lambda} | \Psi_0 \rangle = F_\gamma \langle \Phi_C | \Pi_k (r^{(k)} - r^{(k)}_2) e^{i\xi \ln \lambda} | \Psi_0 \rangle \tag{12}
\]

with \( \Lambda \) defined by Eq.(9) includes the SO terms and also all FSI terms with the accuracy \( 1/R^2 \). The phase factor \( e^{i\xi \ln \lambda} \) is canceled by its conjugated counterpart in the squared amplitude. Employing Eq.(1) we can put \( r^{(k)} = R \), thus presenting

\[
F_x = F_\gamma T_x; \quad T_x = \langle \Phi_x | \Pi_k (1 - t^{(k)}) e^{i\xi} | \Psi_0 \rangle, \tag{13}
\]

with \( t^{(k)} = \frac{p r^{(k)}}{p r^{(k)}} \). Here we omitted the constant factor \( (R\lambda)^{i\xi} \).

The absorption cross section can be defined as the difference between the total cross section and the elastic one. At large \( E \) the sum over the exited states of the FS can be calculated by employing closure approximation. Thus

\[
r(E) = 1 - |\langle \Psi_0 | \Pi_k (1 - t^{(k)}) e^{i\xi} | \Psi_0 \rangle|^2. \tag{14}
\]

This provides

\[
r(E) = 1 - \frac{1}{(1 + \xi^2)^N} = 1 - e^{-N \ln(1 + \xi^2)}. \tag{15}
\]

If the photon energy is so large that \( N\xi^4 \ll 1 \) (for \( C_{60} \) this means that \( E \gg 800 \text{ eV} \)), we find

\[
r(E) = 1 - e^{-N\xi^2}. \tag{16}
\]

In the high energy limit \( N\xi^2 \ll 1 \) the perturbative approach is valid, and \( r(E) \approx N\xi^2 \), thus dropping with \( E \).

IV. APPLICATION TO THE ENDOHEDRAL ATOMS \textit{A@C}_N

There are more or less detailed investigated fullerenes with the number of atoms \( N \): \( 20, 60, 70, 80 \). There are \( N_v = 4N \) valence (collectivized) electrons and \( N_c = 2N \) core
electrons. The latter can be treated as the $1s$ electrons bound by the carbon nuclei (also in the field of the valence electrons, the action of which upon $1s$ is small) with the binding energy $I_c \approx 300 \text{ eV}$. The photoelectron is known to feel the fullerene potential $V \ll 1$. Thus the condition $\xi^2 \ll 1$ enables to use the free propagator presented by Eq.(8). The binding energies $I_{FS}$ of the valence FS electrons satisfy the condition $I_{FS} \ll I_c$. Employing of the closure approximation requires that the photoelectron energy $E$ is large enough to include all important excited states, i.e. $E$ should be much larger than the energy losses $\bar{\varepsilon}$ in the FS. For the energies $\varepsilon$ of the excitation of the FS, which exceed strongly the FS binding energies $I_{FS}$ (e.g. $I_{FS} \approx 7 \text{ eV for } C_{60}$ ) the energy distributions drop as $1/\varepsilon^2$. Thus the values of $\bar{\varepsilon}$ for the valence FS electrons are determined by $I_{FS} \ll \varepsilon \ll E$. They are

$$\bar{\varepsilon} = \frac{\xi^2 N_v}{4R^2} \ln \frac{E}{I_{FS}}.$$  

(17)

As follows from Eq. (17) the closure approximation can be used for $E \gg 50 \text{ eV}$ for all the considered fullerenes. At $E \leq I_c$ only the valence electrons can be knocked out by the FSI. In this region $N_v \xi^2 \geq 3.5$ for $C_{20}$ and is even larger for the fullerenes with larger $N$. Thus, $r(E)$ is described by Eq.(15) with $N = N_v$. At $E = 300 \text{ eV}$ we find $r = 0.97$ for $C_{20}$ and $r = 0.99997$ for $C_{60}$. Hence, the cross section $\sigma_A$ is very close to the cross section of photoionization $\sigma_\gamma$. At $E \sim I_c$ contribution of the core electrons cannot be calculated by employing the closure approximation. However Eq.(15) provides the lower limit for $r(E)$. For $E \gg I_c$ we can employ Eq.(15) with $N = N_v + N_c$. For example, at $E = 1 \text{ keV}$ we find $r = 0.80$ for the fullerene $C_{20}$, while $r = 0.992$ for $C_{60}$. The cross section $\sigma_A$ is again close to $\sigma_\gamma$.

The perturbative behavior requires the energies of the photoelectron to be very large. We find that $N\xi^2 \ll 1$ at $E \gg 1.6 \text{ keV}$ for the fullerene $C_{20}$, and at $E \gg 5 \text{ keV}$ for $C_{60}$. At the energies of dozens of keV the FSI and SO terms are of the same order and the SO terms should be included.

V. SUMMARY

To summarize, we found the energy dependence of the cross section of photoionization of the caged atom accompanied by inelastic processes in the fullerene shell. In the broad intervals of energies this cross section appeared to be close to that of photoionization of
the isolated atom. In other words, the probability that the FS will be excited during photoionization of the caged atom is close to unity. This means that in the related problem of studying the wave function of the outgoing electron in photoionization of the caged atom we cannot consider the FS as just a source of external field. Inelastic processes in the FS are important. The results was obtained by summation of the power series of interaction between the photoelectron and the FS. The technique of summation can be applied for investigation of other objects with two scales of distances. It can be useful for the systems containing a large number of electrons. These can be heavy atoms or big molecules. The photoionization of the inner shell of a heavy atom followed by ionization of an outer one can serve as an example.

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