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Photoionization of 3d electrons of Xe, Cs and Ba endohedral atoms: comparative analyses

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Abstract: We demonstrate rather interesting manifestations of co-existence of resonance features in characteristics of the photoionization of 3d-electrons in Xe, Cs and Ba endohedral atoms. It is shown that for all of the considered atoms the reflection by the fullerene shell of photoelectrons produced by the 3d subshell photoionization affects greatly partial photoionization cross-sections of \(^{3/2}\) and \(^{5/2}\) levels and respective angular anisotropy parameters, both dipole and non-dipole adding to all of them additional maximums and minimums. The results obtained demonstrate distinctive differences between the three atoms. The calculations are performed treating the 3/2 and 5/2 electrons as electrons of different kinds with their spins “up” and “down”. The effect of the C\(_{60}\) shell is accounted for in the frame of the “orange” skin potential model. It is essential that in the considered photon frequency region the presented resonance features are not affected by the C\(_{60}\) polarization.

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1. Introduction

Recently we have published the results of calculations of the photoionization cross-section for 3d electrons in Xe@C\(_{60}\) where along with intra-doublet resonance there exists a strong action of the fullerene C\(_{60}\)potential upon the electron waves that are emitted from both, 3d\(_{3/2}\) and 3d\(_{5/2}\) levels [1]. Existence of these two types of resonances leads to interference patterns of the photoionization cross-section and angular anisotropy parameters, both dipole and non-dipole. The intra-doublet resonance in 3d is located at high enough photon energies, where the role of fullerene polarization by the incoming radiation [2] is entirely inessential.

We know from the studies of intra-doublet resonances in isolated atoms that there is an essential difference in their manifestations in three atoms-neighbors – Xe, Cs, and Ba. While in Xe this resonance is an entirely continuous spectrum phenomenon, it acquires an autoionization nature in Ba. As a result, all characteristics of the photoioniza-
tion process, namely the partial cross-sections and angular anisotropy parameters, are essentially different for 3d in this sequence of elements [3]. Similarly, the photoionization of 3d-electrons in Cs@C$_{60}$ and Ba@C$_{60}$ could be essentially different from that of Xe@C$_{60}$. To investigate this difference is the aim of the present paper. We will present newly obtained data for Cs@C$_{60}$ and Ba@C$_{60}$ and compare it with that for Xe@C$_{60}$. To justify this investigation and put it into the context of the current research in the field under consideration, let us repeat some information that was presented in the introduction to [1]. This will simplify considerably the understanding of the aims and expected results of the current research.

A great deal of attention during the last few years was (and still is) concentrated on photoionization of endohedral atoms. It was demonstrated in a number of papers [4–12] that the fullerene shell adds a prominent resonance structure in the photoionization cross section of endohedral atoms. Although the experimental investigation of A@C$_{60}$ photoionization seems to be very difficult at this moment, it will be inevitably studied in the future. The first measurements for such objects have been already performed[1]. This justifies and stimulates the current efforts of the theorists that are predicting rather non-trivial effects waiting for verification.

The role of C$_{60}$ in A@C$_{60}$ photoionization is manifold. C$_{60}$ acts as a spherical potential resonator that reflects the photoelectron wave coming from an A atom. This leads to interference of out-coming and out-going (reflected) waves and to confinement resonances in the frequency dependence of the photoionization cross sections [8]. These resonances were studied in [4–7] for the example of endohedral Ba and Ca atoms located inside the C$_{60}$ shell. Valence and core photoemission of endohedral atoms A@C$_{60}$ (A=Li, Na, K, Be, Mg, and Ca) and resonances in the total cross section were also studied in [8, 11]. The interference of the photoelectron spherical waves inside the resonator C$_{60}$ affects significantly not only the total cross section but also the angular distribution of photoelectrons. This phenomenon was analysed in [9], where it was shown that the confinement resonances are found also in the frequency dependencies of the dipole and non-dipole parameters of the photoelectron angular distribution. The results of these studies give evidence that the reflection and refraction of the photoelec-

\[ R. Phaneuf, private communication (2007) \]
Photoionization cross-sections of Ba 3d electrons, 3d – ef and 3d – ep transitions.

Figure 4. Photoionization cross-sections of 3d\textsubscript{3/2} and 3d\textsubscript{5/2} electrons in Xe and Xe@C\textsubscript{60}.

Figure 5. Photoionization cross-sections of 3d\textsubscript{3/2} and 3d\textsubscript{5/2} electrons in Cs and Cs@C\textsubscript{60}.

Figure 6. Photoionization cross-sections of 3d\textsubscript{3/2} and 3d\textsubscript{5/2} electrons in Ba and Ba@C\textsubscript{60}.

Electron waves by the potential resonator C\textsubscript{60} is prominent up to 60 – 80 eV of the electron energy. The C\textsubscript{60} shell at some frequencies acts as a dynamical screen that is capable of suppressing or enhancing the incident electromagnetic radiation acting upon the doped atom A [13–15]. This effect is due to dynamical polariza-
dipole angular anisotropy parameter of the collectivized electrons of the fullerene shell. Plasma excitations of these electrons generate the alternating dipole moment. This dipole moment causes the ionization of the electronic shells of the endohedral atom. The screening effects of the C₆₀ shell are particularly strong for incident radiation frequency ω² of about that of the C₆₀ Giant resonance, i.e. 20 – 22 eV, but is noticeable in a much broader region, from the ionization threshold up to 60 – 80 eV.

The resonator and screen effects of the fullerene shell C₆₀ manifest themselves as a resonance structure in the frequency dependencies of the differential and total cross sections of the endohedral atom photoionization. Of interest are the situations when pure atomic resonances are superimposed with resonances connected to reflection and refraction of the atomic photoelectron wave by the C₆₀ shell and with resonances due to plasma excitations of the collectivized electrons of the fullerene shell.

We have studied before [14–16] the effect of C₆₀ upon the most important atomic resonances, the Giant and interference, both in Xe. However, these resonances are located at relatively low photon energies, at about 80–100 eV for the Giant resonance and 30–100 for the interference one. This is why the 5s² photoionization of Xe atom inside C₆₀ shell is affected both by the reflection from the C₆₀ shell and by the fullerene own Giant resonance that is located at about 20 eV.

However, a prominent atomic resonance exists at sufficiently high energies. It was called intra-doublet resonance [3, 17]. It was observed in photoionization of 3d₅/₂ and 3d₃/₂ electrons in Xe [2] and Cs [18] and interpreted and discussed in [3, 17].

Let us recall that the intra-doublet resonance is a result of action of 3d₅/₂ electrons upon 3d₃/₂. The threshold energy of 3d is almost 700 eV in Xe and even a little bit higher in Cs and Ba, i.e. well above the position of the Giant resonance in C₆₀. The interference between the intra-doublet and confinement resonances in Xe@C₆₀ was discussed in [1]. Here we will see similar interference in Cs@C₆₀ and Ba@C₆₀. As in the Xe case, at these energies the action of confinement resonances upon atomic became clear and not disturbed by the virtual or real excitations of C₆₀.

2 Atomic system of units is used in this paper.

repeat its main points here. Let us start with the problem of an isolated atom. In fact, what we present below is not a derivation but an extended explanation of notation used.

![Figure 7](image-url)

Figure 7. Dipole angular anisotropy parameter β(ω) for 3d₃/₂ and 3d₅/₂ electrons of Xe. The solid lines are the result for the endohedral Xe@C₆₀. The corresponding curves near the photoionization threshold are presented in the insets.

The method to treat the interaction of 3d₅/₂ and 3d₃/₂ electrons for an isolated Xe atom as a result of the mutual action of two semi-filled levels with five spin-up(↑) and five spin-down(↓) electrons was presented for the first time in [3]. Then the Random Phase Approximation with Exchange (RPAE) equations for atoms with semi-filled shells (so-called Spin Polarized RPAE or SP RPAE) are solved, as described in e.g. [19]. For semi-filled shell atoms the following relation gives the differential in angle photoionization cross-section by non-polarized light, which is similar to that of closed shell atoms [20] (see also e.g. [21]):

\[
\frac{d\sigma_{\uparrow\downarrow}(\omega)}{d\Omega} = \frac{\sigma_{\uparrow\downarrow}(\omega)}{4\pi} \left[ 1 - \frac{B_{\uparrow\downarrow}^2}{2} P_1(\cos \theta) \right] + \kappa \gamma_{\uparrow\downarrow} P_1(\cos \theta) + \kappa \eta_{\uparrow\downarrow} P_3(\cos \theta), \tag{1}
\]

where κ = ω/c is the photon wave vector, P₁(θ) are the Legendre polynomials, θ is the angle between photon k and photoelectron velocity v, B_{\uparrow\downarrow}(ω) is the dipole, while γ_{\uparrow\downarrow}(ω) and η_{\uparrow\downarrow}(ω) are the so-called non-dipole angular anisotropy parameters, where the arrows ↑↓ relates the corresponding parameters to up and down electrons, respectively.
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Figure 8. Dipole angular anisotropy parameter $\beta(\omega)$ for 3\textit{d}_{3/2} and 3\textit{d}_{5/2} electrons of Cs. The solid lines are the result for the endohedral Cs@C\textsubscript{60}. The corresponding curves near the photoionization threshold are presented in the insets.

Figure 9. Dipole angular anisotropy parameter $\beta(\omega)$ for 3\textit{d}_{3/2} and 3\textit{d}_{5/2} electrons of Ba. The solid lines are the result for the endohedral Ba@C\textsubscript{60}.

Figure 10. Non-dipole angular anisotropy parameter $\gamma(\omega)$ for 3\textit{d}_{3/2} and 3\textit{d}_{5/2} electrons of Xe. The solid lines are the result for the endohedral Xe@C\textsubscript{60}. The corresponding curves near the photoionization threshold are presented in the insets.

Since the sources of linearly polarized radiation are usually used in experiment, another form of the angular distribution is more convenient [22–25] instead of (1):

$$\frac{d\sigma_{\text{IT}l}(\omega)}{d\Omega} = \frac{\sigma_{\text{IT}l}(\omega)}{4\pi} [1 + \beta_{\text{IT}l} P_2(\cos \Theta)] + (\delta_{\text{IT}l} + \gamma_{\text{IT}l} \cos^2 \Theta) \sin \Theta \cos \Phi.$$  \hspace{1cm} (2)

Here $\Theta$ is the polar angle between the vectors of photoelectron's velocity $\mathbf{v}$ and photon's polarization $\mathbf{e}$, while $\Phi$ is the azimuth angle determined by the projection of $\mathbf{v}$ in the plane orthogonal to $\mathbf{e}$ which includes the vector of photon's velocity. The non-dipole parameters in (1) and (2) are connected by the simple relations [21]

$$\gamma_{\text{IT}l} = \kappa \eta_{\text{IT}l}, \quad \frac{\delta_{\text{IT}l}}{5} + \frac{\gamma_{\text{IT}l}}{5} = -\kappa \eta_{\text{IT}l}.$$  \hspace{1cm} (3)

The below-presented results of calculations of non-dipole parameters are obtained using both expressions (1) and (2). There are two possible dipole transitions from sub-shell \textit{l}, namely \textit{l} $\rightarrow$ \textit{l} $\pm$ 1 and three quadrupole transitions \textit{l} $\rightarrow$ \textit{l} $\pm$ 2. Corresponding general expressions for $\beta_{\text{IT}l}$ ($\omega$), $\gamma_{\text{IT}l}$ ($\omega$) and $\eta_{\text{IT}l}$ ($\omega$) are rather complex and content the dipole $d_{\text{IT}l}$ and quadrupole $q_{1\pm2,0}$ matrix elements of photoelectron transitions. In one-electron
Hartree-Fock (HF) approximation these parameters can be presented as [21, 26]:

\[
\beta_{\lambda\mu\nu}(\omega) = \frac{(l+1)(l+2)d_{\mu\nu}^2_{l+1 \rightarrow l} + l(l+1)d_{\mu\nu}^2_{l \rightarrow l-1} - 6(l+1)d_{l+1\mu}d_{l-1\nu}\cos(\delta_{l+1} - \delta_{l-1})}{(2l+1)\left[(l+1)d_{l+1\mu}^2 + ld_{l-1\nu}^2\right]}.
\]  

(4)

It is implied that the indexes \(\uparrow \downarrow\) are added similarly to the parameters \(\gamma_{\lambda\mu}(\omega)\), \(\eta_{\lambda\mu}(\omega)\) and matrix elements \(d_{l\pm1}, q_{l\pm2}\) in (5) and (6):

\[
\begin{align*}
\gamma_{\lambda\mu}(\omega) &= \frac{3}{5}d_{l\mu}^2\left[\frac{l+2}{2l+3}d_{l+1\mu}[3(l+2)d_{l+1\mu}^2 + l(l+1)d_{l-1\mu}^2 - 6l(l+1)d_{l+1\mu}d_{l-1\mu}\cos(\delta_{l+1} - \delta_{l-1}) - lq_1\times\cos(\delta_{l+1} - \delta_{l-1})]\\
&\quad - \frac{l}{2l+1}d_{l-1\mu}[3(l-1)d_{l-2\mu}^2 + (l+1)d_{l+1\mu}^2\cos(\delta_{l+1} - \delta_{l-1}) - (l+1)d_{l-1\mu}\cos(\delta_{l-1} - \delta_{l+1})]\right].
\end{align*}
\]  

(5)
Here $\tilde{d}(k)$ are the photoelectrons' scattering phases; the following relation gives the matrix elements $d_{\pm 1 1}$ in the so-called $r$-form

$$d_{\pm 1 1} = \int_0^\infty P_{nl1} (r) r P_{\pm 1 11} (r) dr,$$

where $P_{nl1} (r)$, $P_{\pm 1 11} (r)$ are the radial so-called Spin-Polarized Hartree-Fock (SP HF) [27] one-electron wave functions of the $nl$ discrete level and $\varepsilon l \pm 1$ continuous spectrum, respectively. The following relation gives the quadrupole matrix elements

$$q_{\pm 2 01} = \frac{1}{2} \int_0^\infty P_{nl1} (r) r^2 P_{\pm 2 01} (r) dr.$$

In order to take into account the Random Phase Approximation with Exchange (RPAE) [26] multi-electron correlations, one has to perform the following substitutions in the expressions for $P_{nl1} (w)$, $y_{nl1} (w)$ and $q_{nl1} (w)$ [21]:

$$d_{i,1} d_{l,-1} \cos (\delta_{i,1} - \delta_{l,-1}) \rightarrow \tilde{D}_{i,1} \tilde{D}_{l,-1} \cos (\delta_{i,1} + \Delta_{i,1} - \delta_{l,-1} - \Delta_{l,-1}) \; .$$

$$d_{i,1} q_{\pm 2 0} \cos (\delta_{i, \pm 2 0} - \delta_{l, \pm 1}) \rightarrow \tilde{D}_{i, \pm 1} \tilde{Q}_{\pm 2 0} \cos (\delta_{i, \pm 2 0} + \Delta_{i, \pm 2 0} - \delta_{l, \pm 1} - \Delta_{l, \pm 1}) \; ,$$

where $\tilde{D}_{i, \pm 1} (w), \tilde{Q}_{\pm 2 0} (w)$, $\Delta_{i, \pm 1}$ and $\Delta_{i, \pm 2 0}$ are the modulus and argument of dipole and quadrupole amplitudes.

The ordinary RPAE equation for the dipole matrix elements is as follows

$$\langle v_2 | D (\omega) | v_1 \rangle = \langle v_2 | d | v_1 \rangle + \sum_{v_3, v_4} \langle v_3 | D (\omega) | v_4 \rangle \left( n_{v_3} - n_{v_4} \right) \langle v_4 v_3 | U | v_1 v_2 \rangle \frac{\epsilon_{v_4} - \epsilon_{v_3} + \omega + i \eta \left( 1 - 2 n_{v_3} \right)}{\eta},$$

where

$$\langle v_1 v_2 | \tilde{U} \tilde{v}_1 \tilde{v}_2 \rangle \equiv \left( \langle v_1 v_2 | \tilde{V} \tilde{v}_1 \tilde{v}_2 \rangle - \langle v_1 v_2 | \tilde{V} \tilde{v}_1 \tilde{v}_2 \rangle \right) \; .$$

Here $\tilde{V} \equiv 1/|r' - r|$ and $v_i$ is the total set of quantum numbers that characterize a HF one-electron state. That includes the principal quantum number (energy), angular momentum, its projection and the projection of the electron spin. The function $n_{v_i}$ (the so-called step-function) is equal to 1 for occupied states and 0 for vacant ones.

For semi-filled shells the RPAE equations are transformed into the following system of equations that can be presented in the matrix form:

$$\left( \tilde{D}_1 (\omega) \; \tilde{D}_1 (\omega) \right) = \left( \tilde{d}_1 (\omega) \; \tilde{d}_1 (\omega) \right) + \left( \tilde{D}_1 (\omega) \; \tilde{D}_1 (\omega) \right) \times \left( \hat{X} \; 0 \right) \times \left( \tilde{X}_1 \; \tilde{V}_1 \right) \; .$$

The dipole matrix elements $D_{i, \pm 1}$ are obtained by solving the radial part of the RPAE equation (12). As to the quadrupole matrix elements $Q_{\pm 2 0}$, they are obtained by solving the radial part of the RPAE equation, similar to (12)

$$\langle v_2 | Q (\omega) | v_1 \rangle = \langle v_2 | \tilde{Q} | v_1 \rangle + \sum_{v_3, v_4} \langle v_3 | Q (\omega) | v_4 \rangle \left( n_{v_3} - n_{v_4} \right) \langle v_4 v_3 | U | v_1 v_2 \rangle \frac{\epsilon_{v_4} - \epsilon_{v_3} + \omega + i \eta \left( 1 - 2 n_{v_3} \right)}{\eta} \; .$$

Here $\tilde{Q} \equiv 1/|r' - r'|$ and $v_i$ is the total set of quantum numbers that characterize a HF one-electron state. That includes the principal quantum number (energy), angular momentum, its projection and the projection of the electron spin. The function $n_{v_i}$ (the so-called step-function) is equal to 1 for occupied states and 0 for vacant ones.
Here in the r-form one has \( \hat{q} = r^2 P_r(\cos \theta) \).

Equations (14, 15) are solved numerically using the procedure discussed at length in [27]. The generalization of (15) for semi-filled shells is similar to (14):

\[
\left( \hat{\Omega}_1(\omega) \right) = \left( \hat{Q}_1(\omega) \hat{Q}_1(\omega) \right) + \left( \hat{Q}_1(\omega) \hat{Q}_1(\omega) \right) \times \left( \begin{array}{cc} \hat{\chi}_1 \hat{\chi}_1 & 0 \\ 0 & \hat{\chi}_1 \hat{\chi}_1 \end{array} \right) \times \left( \begin{array}{c} \hat{V}_1 \\ \hat{V}_1 \\ \hat{U}_1 \end{array} \right).
\]

(16)

where

\[
\hat{\chi}_1(\omega) = \frac{1}{1}(\omega - \hat{H}_\nu) - 1/(\omega + \hat{H}_\nu).
\]

(17)

In (17) \( \hat{H}_\nu \) is the electron – vacancy HF Hamiltonian. Equations (14) and (16) permit treating the electron correlations of 3d_{5/2} and 3d_{3/2} levels, provided they are corrected by adding the factors 6/5 and 4/5 to the up and down states, respectively. These equations were solved in this paper numerically. The cross-sections and angular anisotropy parameters are calculated by using numerical procedures with the codes described in [27].

3. Effect of C\textsubscript{60} fullerene shell

The ionization potentials of the Xe 3d_{5/2} and 3d_{3/2} subshells have the value of about 50 Ry. For this photon energy significantly exceeding the energy of the Giant Resonance of the C\textsubscript{60} collectivised electrons, the screening effects of the fullerene shell do not play any essential role in the photoionization processes of the Xe atom localized inside C\textsubscript{60}. Therefore, the main reason for modifying the photoionization parameters of 3d_{5/2} and 3d_{3/2} subshells of the encapsulated Xe, as compared to the free Xe, are the confinement effects. These effects near the photoionization threshold can be described within the framework of the “orange” skin potential model. According to this model, for small photoelectron energies the real static potential \( V(\omega) \) is equal to its observable value. Addition of potential \( V(\omega) \) at the atomic HF potential leads to a factor \( F_1(\omega) \) in the photoionization amplitudes which depends upon the photoelectron's momentum \( k \) and orbital quantum number \( l \) [29]:

\[
V(r) = -V_0 \delta(r - \hat{R}).
\]

(18)

The parameter \( V_0 \) is determined by the requirement that the binding energy of the extra electron in the negative ion C\textsubscript{60} is equal to its observable value. Addition of potential \( V(\omega) \) to the atomic HF potential leads to a factor \( F_1(\omega) \) in the photoionization amplitudes which depends upon the photoelectron's momentum \( k \) and orbital quantum number \( l \) [29]:

\[
F_1(k) = \cos \eta(k) \left[ 1 - \tan \eta(k) \frac{u_{\epsilon l}(\hat{R})}{\hat{u}_{\epsilon l}(\hat{R})} \right],
\]

(19)

where \( \eta(k) \) are the additional phase shifts due to the fullerene shell potential (18). They are expressed by the following formula:

\[
\tan \eta(k) = \frac{u_{\epsilon l}(\hat{R})}{\hat{u}_{\epsilon l}(\hat{R})} + \frac{k}{2V_0}.
\]

(20)

In these formulas \( u_{\epsilon l}(\hat{R}) \) and \( \hat{u}_{\epsilon l}(\hat{R}) \) are the regular and irregular solutions of the atomic HF equations for a photoelectron with momentum \( k = \sqrt{2\varepsilon} \), where \( \varepsilon \) is the photoelectron energy connected with the photon energy \( \omega \) by the relation \( \varepsilon = \omega - I_A \) with \( I_A \) being the atom A ionization potential.

The applicability of zero-thickness bubble potential for description of the fullerene shell is parametrically justified when the de Broglie wavelength of photoelectron exceeds the effective thickness of the C\textsubscript{60} shell. As it has been shown in [30], consideration of the reasonable finite thickness of the shell has practically no influence on the frequency dependencies of the dipole and non-dipole parameters near the photoionization threshold. This confirms the applicability of the bubble potential model potential. Using Eq. (19), one can obtain the relations for the \( D_{nl\lambda \epsilon}^{AC} \) and \( Q_{nl\lambda \epsilon}^{AC} \) amplitudes of endohedral atom expressed via the respective values for isolated atom. For \( nl \rightarrow \epsilon \lambda' \) transitions they have the following form:

\[
D_{nl\lambda \epsilon}^{AC}(\omega) = F_1(\omega)D_{nl\lambda \epsilon}^{(1)}(\omega),
\]

(21)

and

\[
Q_{nl\lambda \epsilon}^{AC}(\omega) = F_1(\omega)Q_{nl\lambda \epsilon}^{(1)}(\omega).
\]

(22)

For the cross-sections one has

\[
\sigma_{nl\lambda \epsilon}^{AC}(\omega) = F_1^2(\omega)\sigma_{nl\lambda \epsilon}^{(1)}(\omega).
\]

(23)

With these amplitudes, using the expressions (4-6) and performing the substitution (9, 10), we obtain the cross-sections and angular anisotropy parameters for endohedral atoms. While calculating the anisotropy parameters, the cosines of atomic phases differences \( \cos(\delta_l - \delta'_{l'}) \) in formulas (4)-(6) are replaced by \( \cos(\delta_l + \eta_l - \delta'_{l'} - \eta_{l'}) \). Note that factors \( F_1(\omega) \) are different for spin-up(\( \uparrow \)) and spin-down(\( \downarrow \)) photoelectrons and therefore they should be denoted as \( F_{1\uparrow}(\omega) \).

As a result, one has for the dipole angular anisotropy parameter (4), using (9) and (10):
In Figs. 3 the partial cross-sections for \( 3d \rightarrow cf \) were chosen as the same as in the previous papers, e.g. [28]. Some details of calculations and lowing expressions for the non-dipole angular anisotropy parameters:

\[
\beta_{l\ell l_1}(\omega) = \frac{(l + 1)(l + 2)F_{l+1 l_1}^2 \hat{D}_{l+1 l_1}^2 + ll(l - 1)F_{l-1 l_1}^2 \hat{D}_{l-1 l_1}^2 - 6(l + 1)F_{l+1 l_1}F_{l-1 l_1} \hat{D}_{l+1 l_1} \hat{D}_{l-1 l_1} \cos(\hat{\delta}_{l+1} - \hat{\delta}_{l-1})}{(2l + 1)F_{l+1 l_1}^2 \hat{D}_{l+1 l_1}^2 + F_{l-1 l_1}^2 \hat{D}_{l-1 l_1}^2},
\]

where \( \hat{\delta}_l = \delta_l + \Delta_\varepsilon + \eta_\varepsilon \) (see (11)).

From (5) and (6), using (9) and (10), we arrive at the fol-

\[
\gamma_{l\ell l_1}(\omega) = \frac{3}{5(l + 1)F_{l+1 l_1}^2 \hat{D}_{l+1 l_1}^2 + F_{l-1 l_1}^2 \hat{D}_{l-1 l_1}^2} \left\{ \frac{l + 1}{(l + 2)}F_{l-1 l_1} \hat{D}_{l-1 l_1} \cos(\hat{\delta}_{l+2} - \hat{\delta}_{l+1}) - F_{l+1 l_1} \hat{D}_{l+1 l_1} \cos(\hat{\delta}_{l+2} - \hat{\delta}_{l+1}) \right\}
\]

\[
\eta_{l\ell l_1}(\omega) = \frac{3}{5(l + 1)F_{l+1 l_1}^2 \hat{D}_{l+1 l_1}^2 + F_{l-1 l_1}^2 \hat{D}_{l-1 l_1}^2} \left\{ \frac{l + 1}{(l + 2)}F_{l-1 l_1} \hat{D}_{l-1 l_1} \cos(\hat{\delta}_{l+2} - \hat{\delta}_{l+1}) - F_{l+1 l_1} \hat{D}_{l+1 l_1} \cos(\hat{\delta}_{l+2} - \hat{\delta}_{l+1}) \right\}
\]

4. Some details of calculations and their results

Naturally, the \( C_{60} \) parameters in the present calculations were chosen as the same as in the previous papers, e.g. in [28, 29]: \( R = 6.639 \) and \( V_0 = 0.443 \).

In Figs. 1-15 we present our results for the partial cross-sections \( \sigma_{l\ell l_1}(\omega) \) that correspond to \( 3d \rightarrow cf \) and \( 3d \rightarrow ep \) transitions and for the dipole and non-dipole angular anisotropy parameters for Xe, Cs and Ba endohedral atoms. The results for Xe@C\(_{60}\) were already presented in [1], while the data for isolated Xe can be found in [3, 17]. The photoionization cross-section and angular anisotropy parameters for 3d electrons in Cs and Ba were considered in [30].

The partial cross-sections for \( 3d \rightarrow cf \) and \( 3d \rightarrow ep \) transitions in Xe, Cs and Ba are presented in Figs. 1, 2, 3, in its upper and lower parts, respectively. The solid lines in these figures present the 3d\(_{5/2}\) cross-sections while the dashed line stands for the 3d\(_{3/2}\) cross-sections. The contribution of the transition \( 3d \rightarrow ep \) exceeds that of \( 3d \rightarrow cf \) by almost two orders of magnitude. An additional maximum in both transitions from the 3d\(_{3/2}\) level is due to action of the electrons on the 3d\(_{3/2}\) level. Since the ionization potentials of the "up" and "down" levels are different, the corresponding curves are shifted relative to each other along the axis of photon energy. The main maxima in the \( 3d \rightarrow cf \) transition are increasing on the way from Xe to Ba. In Ba it is a result of autoionization of the discrete excitation of 3d\(_{1/2}\) electrons.

Since the partial cross-section corresponding to the \( 3d \rightarrow ep \) transition is much smaller than the \( 3d \rightarrow cf \) partial cross-section, when considering the effect of the fullerene
shell on the $\text{Xe@C}_{60}$ photoionization cross-section we concentrate only on the main $3d \to ef$ electron transition.

In Figs. 4, 5, 6 we present the partial photoionization cross-section of $3d_{3/2}$ and $3d_{5/2}$ levels in $\text{Xe@C}_{60}$, $\text{Cs@C}_{60}$, and $\text{Ba@C}_{60}$. The solid lines in these figures depict the results for endohedral atoms, while the dashed line presents the same for isolated atoms. The upper part of Figs. 4, 5, 6 demonstrates the effect of $\text{C}_{60}$ upon the photoionization cross-section of the $3d_{3/2}$ electrons, while the lower parts present the results for $3d_{5/2}$ electrons. The main maximum in $\text{Xe@C}_{60}$ and $\text{Cs@C}_{60}$ remains almost the same, while in $\text{Ba@C}_{60}$ it is considerably smaller. All cross-sections acquire a prominent oscillating structure around the background curves, due to reflection of the $ef$ photoelectron wave by the $\text{C}_{60}$ shell.

Note that the additional maximum that appears due to the $3d_{3/2}$ action is prominently altered under the influence of the $\text{C}_{60}$ shell. Several additional secondary maxima are created. The complex oscillating structure makes the cross-sections of $5/2$ and $3/2$ electrons in $\text{Xe@C}_{60}$ similar. To a lesser extent this similarity takes place also in $\text{Cs@C}_{60}$. In $\text{Ba@C}_{60}$ the amplitude of oscillations is even bigger for the $3/2$ cross-section.

The amplitude factor Eq. (18) is defined by the values of the photoelectron wave functions with the “up” and “down” spins at $r = R$. Since we deal with the completely filled subshell the role of the spin effects in the wave function behaviour is small. Hence the continuum wave functions for the “up” and “down” spins at this point are almost equal. So, the amplitude factors $F_f(k)$ for the $3/2$ and $5/2$ levels are similar to each other. However, since the background cross-sections for the isolated $\text{Xe}$, $\text{Cs}$ and $\text{Ba}$ atoms are significantly different, the interference of $3/2$ and $5/2$ intra-doublet resonances and confinement resonances leads to essentially different cross-sections for $\text{Xe@C}_{60}$, $\text{Cs@C}_{60}$, and $\text{Ba@C}_{60}$ compounds, which we see in Figs. 4, 5, 6.

In Figs. 7, 8, 9 we depict the dipole angular anisotropy parameter $b(\omega)$ for the endohedrals $\text{Xe@C}_{60}$, $\text{Cs@C}_{60}$, and $\text{Ba@C}_{60}$ in the comparison of the isolated atoms $\text{Xe}$, $\text{Cs}$ and $\text{Ba}$, respectively. Impressive modifications are seen. The solid lines in these figures depict the results for endohedral atoms, while the dashed line presents the same for isolated atoms. The upper part of Figs. 7, 8, 9 demonstrates the effect of $\text{C}_{60}$ upon the photoionization cross-section of the $3d_{5/2}$ electrons. The lower part presents the results for $3d_{3/2}$ electrons. Although being an order of magnitude smaller than the $3d \to ef$ amplitude, the $3d \to ep$ amplitude is much more important in the angular distributions of photoelectrons than in partial cross-sections. It is seen that the action of $3d_{3/2}$ electrons leads to an extra maximum in the $3d_{5/2}$ cross-section.

It is interesting but perhaps incidental that the position of the main maxima in free $\text{Xe}$ and $\text{Xe@C}_{60}$, just as in free $\text{Cs}$ and $\text{Cs@C}_{60}$, almost coincide but the entire endohedral atom structure is much more complex. Figs. 7, 8, 9 demonstrates the noticeable modifications in the dipole angular anisotropy parameter $b(\omega)$. However, the general behavior of $b(\omega)$, which in essence is a ratio (see (4) and (24)) of dipole matrix elements, is for endohedrals and for isolated atoms similar. Within the whole range of photon energy under consideration the curves $b(\omega)$ for the endohedral atoms $\text{Xe@C}_{60}$, $\text{Cs@C}_{60}$, and $\text{Ba@C}_{60}$ oscillate relative to the atomic curve. The fullerene shell especially vividly changes the frequency dependence of the parameter near the photoeffect threshold, where additional structure appears that is most impressive for $\text{Xe@C}_{60}$ and ionization of the $3/2$ electrons in $\text{Ba}$. The additional maximum of the curve $b_{5/2}(\omega)$ for photon energy $\omega \approx 51.5$ Ry exceeds by almost one and a half the maximum of the curve $b_{3/2}(\omega)$. Note that reflection by $\text{C}_{60}$ generates in $b_{3/2}(\omega)$ a maximum similar to the maximum caused by intra-doublet correlations in $b_{5/2}(\omega)$. Some additional structure is created due to confinement of $\text{Xe}$ inside $\text{C}_{60}$.

Figs. 10, 11, 12 and Figs. 13, 14, 15 present the non-dipole parameters $g(\omega)$ and $\delta(\omega)$, respectively, determined by using equations (5), (6), (25), (26) and (3). The dashed lines on these figures give the data for isolated atoms, while the solid lines correspond to the endohedral atoms $\text{Xe@C}_{60}$, $\text{Cs@C}_{60}$, and $\text{Ba@C}_{60}$. As before, the upper curves correspond to ionization of the $5/2$, while the lower ones to the $3/2$ electrons. In Figs. 10-15 we also observe the oscillations of the frequency dependence parameters which are especially vivid near the process threshold where the behavior of the background parameters for the $3/2$ and $5/2$ electrons are different as well. Interplay of intra-doublet resonances with confinement ones leads to strong interference phenomena that we can see in these figures. As a result of this interplay, the shapes of $\gamma_{5/2}(\omega)(\delta_{5/2}(\omega))$, and $\gamma_{3/2}(\omega)(\delta_{3/2}(\omega))$ for $\text{Xe@C}_{60}$ and $\text{Cs@C}_{60}$ (Figs. 10 and 11, Figs. 13 and 14), respectively, became closer than in isolated atoms where $\gamma_{5/2}(\omega)(\delta_{5/2}(\omega))$ has an additional maximum, while $\gamma_{3/2}(\omega)(\delta_{3/2}(\omega))$ has not.

Entirely, we see that the presence of the $\text{C}_{60}$ shell leads to prominent extra additional resonance structure in all the characteristics of the photoionization of $\text{Xe@C}_{60}$, $\text{Cs@C}_{60}$, and $\text{Ba@C}_{60}$ $3d_{5/2,3/2}$ electrons. It justifies studies of the photoionization of here-considered endohedrals in the future. It has been discovered recently [18] that in accord with predictions in [3] for Cs and Ba the intra-doublet reso-
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Figure 13. Non-dipole angular anisotropy parameter $\delta'(\omega)$ for 3d_{5/2} and 3d_{3/2} electrons of Xe. The solid lines are the result for the endohedral Xe@C_{60}. The corresponding curves near the photoionization threshold are presented in the insets.

Figure 14. Non-dipole angular anisotropy parameter $\delta'(\omega)$ for 3d_{5/2} and 3d_{3/2} electrons of Cs. The solid lines are the result for the endohedral Cs@C_{60}.

Figure 15. Non-dipole angular anisotropy parameter $\delta'(\omega)$ for 3d_{5/2} and 3d_{3/2} electrons of Ba. The solid lines are the result for the endohedral Ba@C_{60}.

A qualitative confirmation of the above-presented results of calculations, according to which the resonance structure in the 3d_{5/2} and 3d_{3/2} cross-sections of Cs@C_{60} and Ba@C_{60} is even stronger than in Xe@C_{60}.

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