Non-dipole angular anisotropy parameters of photoelectrons from semi-filled shell atoms

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Abstract
We present the results of calculations of outer and next to outer shell non-dipole angular anisotropy parameters of photoelectrons for semi-filled shell atoms in the Hartree–Fock (HF) one-electron approximation and in the frame of the spin polarized random phase approximation with exchange (SP RPAE) which takes into account inter-electron correlations. We demonstrate for the first time that this characteristic of the photoionization process is essentially sensitive to whether the photoelectron has the same or opposite spin orientation to that of the semi-filled shell.

1. Introductory remarks
The non-dipole parameters of the photoelectron angular distribution are relatively small quantities, in general smaller than the dipole parameter by the factor $v_i/c \ll 1$, where $v_i$ is the mean speed of the ionized electron and $c$ is the speed of light. However, as a result of the construction of high intensity continuous spectrum radiation devices, namely synchrotrons and storage rings, the non-dipole parameters have become measurable quantities.

These parameters are of great interest including information not only on dipole transition matrix elements but also on quadrupole ones. Therefore they can demonstrate the existence of quadrupole resonances that are invisible in the absolute cross-section and shed light on the behaviour of photoelectron scattering phases that corresponds not only to $l \rightarrow l \pm 1$ but also to $l \rightarrow l, l \pm 2$ transitions, where $l$ is the angular momentum of the ionized electron.

The expression for the differential in energy cross-section has been well known for a long time \cite{1–3}, as well as the concrete expressions for the dipole parameter $\beta_{nl}(\omega)$ \cite{3, 4} and non-dipole parameters $\gamma_{nl}(\omega)$ and $\eta_{nl}(\omega)$ \cite{4, 5} for photoionization of an $nl$ subshell (where $\omega$ is the photon frequency\textsuperscript{3}) in the frame of the one-electron Hartree–Fock (HF) approximation and the random phase approximation with exchange (RPAE), which takes into account the most important part of the inter-electron correlations.

Note that we use the atomic system of units $e = m_e = \hbar = 1$ in this paper.

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Extensive calculations of non-dipole parameters were performed mainly for closed shell ions and atoms \([6, 7]\). Special attention was given to noble gases \([8]\) and negative ions \([9]\). There exist several papers dedicated to the results of non-dipole parameter measurements (see, e.g., \([10–12]\)). However, almost nothing has been done in this direction for open shell atoms, particularly with accounting for the role of electron correlations. The latter is especially important in these atoms.

The general consideration of open shell atoms in the frame of RPAE is rather complex. But it was demonstrated quite a while ago that semi-filled shell atoms, being in one sense open shell atoms, can be treated in the RPAE frame as closed shell atoms with two types, namely ‘up’ and ‘down’ electrons, thus labelled according to their spin direction. The corresponding generalization of RPAE can be found in \([13]\).

2. Main formulae

In this section we will present the main formulae used in this paper. Since they are known, we will limit ourselves to the final results only, omitting the corresponding derivations. The following relation gives the differential in angle photoionization cross-section by non-polarized light \([4]\):

\[
\frac{d\sigma_{nl}(\omega)}{d\Omega} = \frac{\sigma_{nl}(\omega)}{4\pi} \left[ 1 - \frac{\beta_{nl}(\omega)}{2} P_2(\cos \theta) + \kappa \gamma_{nl}(\omega) P_1(\cos \theta) + \kappa \eta_{nl}(\omega) P_3(\cos \theta) \right] .
\]

(1)

where \(\kappa = \omega/c\), \(P_{1,2,3}(\cos \theta)\) are the Legendre polynomials, \(\theta\) is the angle between the photon and photoelectron momenta, \(\beta_{nl}(\omega)\) is the dipole, while \(\gamma_{nl}(\omega)\) and \(\eta_{nl}(\omega)\) are so-called non-dipole angular anisotropy parameters.

In experiments, sources of linearly polarized radiation are usually used. In this case instead of (1) another form of angular distribution is more convenient \([14, 15]\):

\[
\frac{d\sigma_{nl}(\omega)}{d\Omega} = \frac{\sigma_{nl}(\omega)}{4\pi} \left[ 1 + \beta_{nl}(\omega) P_2(\cos \vartheta) + \left[ \delta_{nl}^C(\omega) + \gamma_{nl}^C(\omega) \cos^2 \vartheta \right] \sin \vartheta \cos \Phi \right].
\]

(2)

Here \(\vartheta\) is the polar angle between the vectors of the photoelectron velocity \(\vec{v}\) and the photon polarization \(\vec{e}\), while \(\Phi\) is the azimuth angle determined by the projection of \(\vec{v}\) in the plane orthogonal to \(\vec{e}\) that includes the vector of the photon velocity. The non-dipole parameters in (1) and (2) are connected by simple relations \([16]\).

\[
\gamma_{nl}^C/5 + \delta_{nl}^C = \kappa \gamma_{nl}, \quad \gamma_{nl}^C/5 = -\kappa \eta_{nl}.
\]

(3)

The results of calculations of non-dipole parameters presented below are obtained using both expressions, namely (1) and (2).

There are two possible dipole transitions from subshell \(l\), namely \(l \rightarrow l \pm 1\) and three quadrupole transitions \(l \rightarrow l \pm 2, l\). The corresponding general expressions for \(\beta_{nl}(\omega), \gamma_{nl}(\omega)\) and \(\eta_{nl}(\omega)\) are rather complex and are expressed via the matrix elements of these transitions. In the one-electron Hartree–Fock approximation they can be presented as \([3, 8]\)

\[
\beta_{nl}(\omega) = \frac{1}{(2l + 1)[(l + 1)d_{l+1}^2 + ld_{l-1}^2]} \left[ (l + 2)(l + 1)d_{l+1}^2 + l(l - 1)d_{l-1}^2 - 6(l + 1)ld_{l+1}d_{l-1} \right. \\
\left. \times \cos(\delta_{l+1} - \delta_{l-1}) \right] .
\]

(4)

\[
\gamma_{nl}(\omega) = \frac{3}{5[l d_{l-1}^2 + (l + 1)d_{l+1}^2]} \left[ \frac{l + 1}{2l + 3} \left[ 3(l + 2)q_{l+2}d_{l+1} \cos(\delta_{l+2} - \delta_{l+1}) - lq_{l}d_{l+1} \right. \\
\left. \times \cos(\delta_{l+2} - \delta_{l+1}) \right] - \frac{l}{2l + 1} \left[ 3(l - 1)q_{l-2}d_{l-1} \cos(\delta_{l-2} - \delta_{l-1}) \\
\right. \\
\left. - (l + 1)q_{l}d_{l-1} \cos(\delta_{l} - \delta_{l-1}) \right] \right] .
\]

(5)
\[ \eta_{nl}(\omega) = \frac{3}{5} \left[ d_{l+1} d_{l-1} + (l + 1) d_{l+1}^2 \right] \left\{ (l + 1)(l + 2) q_{l+2} [5 d_{l-1} \cos(\delta_{l+2} - \delta_{l-1}) - (l + 3) d_{l+1} \cos(\delta_{l+2} - \delta_{l-1})] - \frac{(l - 1) l}{(2l + 1)(2l + 3)} q_{l-2} \right. \\
\times [5(l + 1) d_{l+1} \cos(\delta_{l-2} - \delta_{l+1}) - (l - 2) d_{l-1} \cos(\delta_{l-2} - \delta_{l+1})] + 2 \frac{l(l + 1)}{(2l - 1)(2l + 3)} q_{l+1} [(l + 2) d_{l+1} \cos(\delta_{l+1} - \delta_{l-1}) - (l - 1) d_{l-1} \cos(\delta_{l} - \delta_{l-1})] \right\}, \] (6)

Here \( \delta_l \) are photoelectron scattering phases; the following relation gives the matrix elements \( d_{\pm 1} \) in the so-called \( r \)-form,

\[ d_{\pm 1} \equiv \int_0^\infty P_{nl}(r) r P_{nl \pm 1}(r) \, dr, \] (7)

where \( P_{nl}(r) \), \( P_{nl \pm 1}(r) \) are the radial Hartree–Fock one-electron wavefunctions of the \( nl \) discrete level and \( n'l' \) in continuous spectrum, respectively. The following relation gives the quadrupole matrix elements:

\[ q_{l \pm 2,0} = \frac{1}{2} \int_0^\infty P_{nl}(r) r^2 P_{nl \pm 2,0}(r) \, dr. \] (8)

In order to take into account the random phase approximation with exchange (RPAE) [3, 17] multi-electron correlations, one has to perform the following substitutions in the expressions for \( \beta_{nl}(\omega) \), \( \gamma_{nl}(\omega) \) and \( \eta_{nl}(\omega) \) [8):

\[ d_{l+1} d_{l-1} \cos(\delta_{l+1} - \delta_{l-1}) \rightarrow \{ (\text{Re} D_{l+1} \text{Re} D_{l-1} + \text{Im} D_{l+1} \text{Im} D_{l-1}) \cos(\delta_{l+1} - \delta_{l-1}) \} \]

\[ - (\text{Re} D_{l+1} \text{Im} D_{l-1} - \text{Im} D_{l+1} \text{Re} D_{l-1}) \sin(\delta_{l+1} - \delta_{l-1}) \} \}

\[ d_{l \pm 1} q_{l \pm 2,0} \cos(\delta_{l \pm 2} - \delta_{l \pm 1}) \rightarrow \{ (\text{Re} D_{l \pm 1} \text{Re} Q_{l \pm 2,0} + \text{Im} D_{l \pm 1} \text{Im} Q_{l \pm 2,0}) \cos(\delta_{l \pm 2} - \delta_{l \pm 1}) \} \]

\[ - (\text{Re} D_{l \pm 1} \text{Im} Q_{l \pm 2,0} - \text{Im} D_{l \pm 1} \text{Re} Q_{l \pm 2,0}) \sin(\delta_{l \pm 2} - \delta_{l \pm 1}) \} \}

\[ d_{l \pm 1}^2 \rightarrow \text{Re} D_{l \pm 1}^2 + \text{Im} D_{l \pm 1}^2. \] (9)

The following is the RPAE equation for the dipole matrix elements,

\[ \langle v_2 \mid D(\omega) \mid v_1 \rangle = \langle v_2 \mid d \mid v_1 \rangle + \sum_{i_1, i_2} \frac{\langle v_1 \mid D (\omega) \mid v_i \rangle \langle n_{i_1} - n_{i_2} \rangle \langle v_1 v_2 \mid U \mid v_3 v_4 \rangle}{\epsilon_{i_1} - \epsilon_{i_2} + \omega + i \eta (1 - 2n_{i_2})}, \] (11)

where

\[ \langle v_1 v_2 \mid \hat{U} \mid v_3 v_4 \rangle = \langle v_1 v_2 \mid \hat{V} \mid v_3 v_4 \rangle = \langle v_1 v_2 \mid \hat{V} \mid v_3' v_4' \rangle. \] (12)

Here \( \hat{V} \equiv 1/|\vec{r} - \vec{r}'| \) and \( v_i \) is the total set of quantum numbers that characterize a HIF one-electron state on discrete (continuum) levels. This includes principal quantum number (energy), angular momentum, its projection and the projection of the spin. The function \( n_{i_1} \) (the so-called step-function) is equal to 1 for occupied and 0 for vacant states.

The dipole matrix elements \( D_{l \pm 1} \) are obtained by solving the radial part of the RPAE equation (11). As to the quadrupole matrix elements \( Q_{l \pm 2,0} \), they are obtained by solving the radial part of the RPAE equation, similar to (11),

\[ \langle v_2 \mid Q(\omega) \mid v_1 \rangle = \langle v_2 \mid q \mid v_1 \rangle + \sum_{i_1, i_2} \frac{\langle v_1 \mid Q (\omega) \mid v_i \rangle \langle n_{i_1} - n_{i_2} \rangle \langle v_1 v_2 \mid U \mid v_3 v_4 \rangle}{\epsilon_{i_1} - \epsilon_{i_2} + \omega + i \eta (1 - 2n_{i_2})}. \] (13)

Here in \( r \)-form one has \( \hat{q} = r^2 P_2 (\cos \theta) \). Equations (11) and (13) are solved numerically using the procedure discussed at length in [17].
Expressions (4)–(6) are essentially simplified for \(s\)-subshells that are the main subject of this paper. They become equal to
\[
\begin{align*}
\beta_{\sigma 0}(\omega) &= 2 \\
\gamma_{\sigma 0}(\omega) &= -\eta_{\sigma 0}(\omega) = \frac{6}{5d^2_l}d_1q_2 \cos(\delta_d - \delta_p), \\
\gamma_{\sigma 0}^c(\omega) &= \frac{6\kappa}{d^2_l}d_1q_2 \cos(\delta_d - \delta_p),
\end{align*}
\]where \(d_1(q_2)\) are the dipole (quadrupole) matrix elements \(\langle \sigma \, p(d)(q) \mid ns \rangle\), and \(\delta_{p,d}\) are the scattering phases of the \(p(d)\)-photoelectron. In RPAE the expression for \(\gamma_{\sigma 0}(\omega) = -\eta_{\sigma 0}(\omega)\) can be obtained from (10) by substituting
\[
d_1q_2 \cos(\delta_2 - \delta_1) \to [(\text{Re} \, D_1 \, \text{Re} \, Q_2 + \text{Im} \, D_1 \, \text{Im} \, Q_2) \cos(\delta_2 - \delta_1)]
- (\text{Re} \, D_1 \, \text{Im} \, Q_2 - \text{Im} \, D_1 \, \text{Re} \, Q_2) \sin(\delta_2 - \delta_1)].
\]
\(d^2_l \to \text{Re} \, D^2_1 + \text{Im} \, D^2_2\).

In this paper we investigate semi-filled shell atoms. As was demonstrated in [13], they can be treated as objects that consist of two types of different particles, namely ‘up’ and ‘down’ electrons, according to their spin projection, \(\uparrow\) and \(\downarrow\), with no exchange between them. In such a case the ordinary HF approximation has to be replaced by spin-polarized Hartree–Fock (SP HF) and RPAE has to be replaced by SP RPAE. Most conveniently this can be done in the operator, symbolic form.

The dipole RPAE equation (11) in the operator form looks like
\[
\hat{D}(\omega) = d + \hat{D}(\omega)\hat{\chi}(\omega)\hat{U},
\]
where the so-called electron–vacancy propagator \(\hat{\chi}(\omega)\) is given by the following relation:
\[
\hat{\chi}(\omega) = \frac{1}{\omega - \hat{H}_{\text{ev}}} - \frac{1}{\omega + \hat{H}_{\text{ev}}},
\]
where \(\hat{H}_{\text{ev}}\) is the electron–vacancy Hartree–Fock Hamiltonian.

As a result of the difference of ‘up’ and ‘down’ states, all the subshells become separated into two levels. For semi-filled atoms all these levels are totally filled with ‘up’, \(\uparrow\), and ‘down’, \(\downarrow\), electrons [13]. The corresponding generalization of RPAE is easy to achieve using instead of (18) the following equation that has a matrix form:
\[
(\hat{D}_1(\omega)\hat{D}_2(\omega)) = (\hat{d}_1(\omega)\hat{d}_1(\omega)) + (\hat{D}_1(\omega)\hat{D}_1(\omega)) \times \begin{pmatrix} \hat{\chi}_{\uparrow\uparrow} & 0 \\ 0 & \hat{\chi}_{\downarrow\downarrow} \end{pmatrix} \times \begin{pmatrix} \hat{V}_{\uparrow\uparrow} & \hat{V}_{\uparrow\downarrow} \\ \hat{V}_{\downarrow\uparrow} & \hat{V}_{\downarrow\downarrow} \end{pmatrix}.
\]
Here the arrows \(\uparrow(\downarrow)\) denote the one-electron functions related to ‘up’ (‘down’) electrons, respectively. Note the presence of the pure direct (without exchange) interaction \(\hat{V}\) between electron–vacancy pairs created by exciting either ‘up’ or ‘down’ electrons.

The generalization of (13) is similar:
\[
(\hat{Q}_1(\omega)\hat{Q}_2(\omega)) = (\hat{q}_1(\omega)\hat{q}_1(\omega)) + (\hat{Q}_1(\omega)\hat{Q}_1(\omega)) \times \begin{pmatrix} \hat{\chi}_{\uparrow\uparrow} & 0 \\ 0 & \hat{\chi}_{\downarrow\downarrow} \end{pmatrix} \times \begin{pmatrix} \hat{V}_{\uparrow\uparrow} & \hat{V}_{\uparrow\downarrow} \\ \hat{V}_{\downarrow\uparrow} & \hat{V}_{\downarrow\downarrow} \end{pmatrix}.
\]
Since ‘up’ or ‘down’ electrons are different, the angular distribution for them is also different, thus acquiring along with \(nl\) an additional lower index \(\uparrow(\downarrow)\). The same is correct for the absolute cross-section that becomes \(\sigma_{\alpha}(\omega)\) and angular anisotropy parameters that become \(\beta_{\alpha}(\omega)\), \(\gamma_{\alpha}(\omega)\) and \(\eta_{\alpha}(\omega)\). They are all expressed via spin-polarized matrix elements \(d_{\uparrow\uparrow}, q_{\uparrow\uparrow}\) and \(d_{\downarrow\downarrow}, q_{\downarrow\downarrow}\) determined
by relations similar to (7) and (8) which correspond to ‘up’ or ‘down’ wavefunctions:

\[ d_{\pm\pm(\downarrow)} = \int_0^{\infty} P_{n\pm\uparrow(\downarrow)}(r) r^2 P_{d\pm\pm(\downarrow)}(r) \, dr, \]
\[ q_{\pm\pm(\downarrow)} = \frac{1}{2} \int_0^{\infty} P_{n\pm\uparrow(\downarrow)}(r) r^3 P_{d\pm\pm(\downarrow)}(r) \, dr. \]

To obtain \( \beta_{n\pm\uparrow(\downarrow)}(\omega) \), \( \gamma_{n\pm\uparrow(\downarrow)}(\omega) \) and \( \eta_{n\pm\uparrow(\downarrow)}(\omega) \) in SP RPAE, substitutions similar to (10) have to be done, using solutions (20) and (21).

3. Results of calculations

We present here results of calculations of non-dipole photoelectron angular anisotropy parameters for outer s-electrons along with the nearest semi-filled subshells of Li \((^2S_1/2)\), P \((^4S_{3/2})\), Cr \((^6S_{5/2})\), Cr* \((^6S_{5/2})\), Mn \((^6S_{5/2})\) and Eu \((^8S_{7/2})\).

The level structure of these atoms looks as follows:

- Li: 1s \( \uparrow \); 2s \( \uparrow \);
- P: 1s \( \uparrow \); 2s \( \uparrow \); 2p \( \uparrow \); 3s \( \uparrow \); 3p \( \uparrow \);
- Cr: 1s \( \uparrow \); 2s \( \uparrow \); 2p \( \uparrow \); 3s \( \uparrow \); 3p \( \uparrow \); 3d \( \uparrow \); 4s \( \uparrow \);
- Cr*: 1s \( \uparrow \); 2s \( \uparrow \); 2p \( \uparrow \); 3s \( \uparrow \); 3p \( \uparrow \); 3d \( \uparrow \); 4s \( \uparrow \);
- Mn: 1s \( \uparrow \); 2s \( \uparrow \); 2p \( \uparrow \); 3s \( \uparrow \); 3p \( \uparrow \); 3d \( \uparrow \); 4s \( \uparrow \);
- Eu: 1s \( \uparrow \); 2s \( \uparrow \); 2p \( \uparrow \); 3s \( \uparrow \); 3p \( \uparrow \); 3d \( \uparrow \); 4s \( \uparrow \); 4p \( \uparrow \); 4d \( \uparrow \); 5s \( \uparrow \); 5p \( \uparrow \); 6s \( \uparrow \).  

With a single line we denote the semi-filled subshell that acts upon outer s-electrons. A double line emphasizes the latter.

The angular anisotropy parameters \( \gamma_{C}(\omega) \) in the HF approximation were obtained using (16), while the RPAE corrections were included using substitution (17). For all but Li atoms, the non-dipole parameters are dependent upon the term of the ion in the final state. This term corresponds to a spin that is smaller by 1/2 than the spin of the initial state if an ‘up’ electron is eliminated, and bigger by 1/2 if a ‘down’ electron is eliminated.

In these notations ‘down’ means the final state for s-electrons photoionization for P – 5S2, Cr* – 6S5/2, Mn – 7S5/2, Eu – 8S4, while ‘up’ means for P – 3S1, Cr – 6S5/2, Mn – 5S2, Eu – 7S3. Note that for Cr and Cr* the final state term after outer s-electron photoionization remains, naturally, the same.

Figure 1 for Li \( \gamma_{C} \) shows no influence of the final state term. In contrast, the onset of the 1s threshold affects \( \gamma_{C} \), also presented in figure 1, leading to a prominent maximum at about 50 eV. The situation in P, as is seen from figure 2, is completely different: \( \gamma_{C} \) and \( \gamma_{C} \) have not too much in common, particularly close to respective rather different thresholds. In both of them the effect of 3p-electrons is quite impressive. As depicted in figure 3, \( \gamma_{C} \) has a strong maximum at 31 eV, while \( \delta_{C} \), being by a factor of ten smaller than \( \gamma_{C} \), has at 32 eV a deep minimum. Note that \( \gamma_{C} \) and \( \delta_{C} \) are almost unaffected by electron correlations.

As seen from figure 4 for Cr, the difference between \( \gamma_{C} \) and \( \gamma_{C} \) is big and impressive, particularly close to thresholds that are almost the same for the ‘up’ and ‘down’ cases. The role of electron correlations in \( \gamma_{C} \) is negligible according to figure 5. As is seen from respective figures 6 and 7, the situation is almost the same in Mn.
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Figure 1. Non-dipole parameter $\gamma_c$ for 1s- and 2s-electrons in Li. The solid line is SP RPAE and the dashed line is HF results.

Figure 2. Non-dipole parameter $\gamma^C$ for 3s-electrons in P. Here ‘down’ means $^5S_2$, ‘up’ – $^3S_1$, final state term of the ion. SP RPAE takes into account the interaction of 3s with 3p-electrons. The solid line is SP RPAE, the dashed line is HF results.

In Eu, as presented in figure 8, the difference between $\gamma_{6s\downarrow}^C$ and $\gamma_{6s\uparrow}^C$ is very big within the 10 eV region near the threshold. At higher energies they are close to each other and very small.

Entirely, we detect a strong dependence of the non-dipole angular anisotropy parameters for outer s-electrons in semi-filled shell atoms upon the term of the residual ion. This effect...
Figure 3. Non-dipole parameters for 3p ↑-electrons in P. The solid line is SP RPAE and the dashed line is HF results.

Figure 4. Non-dipole parameter $\gamma^c$ for the 4s-electron in the excited and ground states of Cr with the terms $^5S_2$ and $^7S_3$, respectively. SP RPAE takes into account the interaction of 4s with 3d-electrons. The solid line is SP RPAE and the dashed line is HF results.
Figure 5. Non-dipole parameters for 3d $\uparrow$-electrons in Cr.

Figure 6. Non-dipole parameter $\gamma_c^d$ for 4s-electrons in Mn. SP RPAE takes into account the interaction of 4s with 3d-electrons. The solid line is SP RPAE and the dashed line is HF results.
Figure 7. Non-dipole parameters for 3d $\uparrow$-electrons in Mn.

Figure 8. Non-dipole parameter $\gamma^c$ for 6s-electrons in Eu. SP RPAE takes into account the interaction of 6s with 4f-electrons. The solid line is SP RPAE and the dashed line is HF results.
can be detected experimentally since the term-dependence shows up in rather prominent corrections that are within the already achieved experimental accuracy (see, e.g., [10]). Note that in spite of the fact that the non-dipole anisotropy parameter is usually for low enough photon energies much smaller than the dipole one (equal to 2 for s-subshells), it is quite measurable experimentally almost from the threshold even for He [19].

Until now, the main attention in studies of non-dipole parameters was concentrated mainly on noble gas atoms, particularly on their inner and intermediate subshells. We do hope that the results presented in this paper will stimulate the efforts of experimentalists who will pay more attention to the investigation of the non-dipole parameters of the outer subshells not only of noble gas atoms, but of other atoms as well.

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