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On the Polarization Dependence of Two-Photon Processes including X-ray Scattering

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Abstract: The polarization dependence of the cross sections of two-photon transitions including X-ray scattering was analyzed. We developed the regular approach to the derivation of the polarization parameters of photoprocesses. Our approach is based on the tensor representation of the photon density matrix, which is written in terms of the unit vectors directed along the major axis of the polarization ellipse ($\hat{\epsilon}$) and the photon propagation ($\hat{\mathbf{k}}$). Explicit expressions for the product of two photon density matrices were derived. As an example, the parametrization of the polarization dependence of the X-ray scattering by closed-shell atoms is given both in terms of (i) scalar products of photon vectors $\hat{\epsilon}_{1,2}$, $\hat{\mathbf{k}}_{1,2}$ and (ii) X-ray Stokes parameters. We discuss the applicability of the atomic scattering for the measurement of the polarization of X-rays.

Keywords: X-rays; scattering; polarization; circular dichroism; angular distributions

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1. Introduction

The process of X- and γ-ray scattering by atomic targets is of great importance in many scientific and technological applications, including non-destructive testing of materials, industrial radiation processing, etc. [1].The Rayleigh scattering of hard X-rays has been intensively studied for many decades (see, e.g., the review article [2]), and a large amount of information on the atomic X-ray Rayleigh scattering is available [2–5]. Recently, this process has attracted interest as a potential tool for measuring the polarization properties of X-rays [6–9]. Currently, the polarization of VUV and XUV radiation is analyzed using gas phase ionization detectors [10,11]. However, for larger photon energies, the photoionization cross section rapidly decreases, so that the efficiency of gas ionization detectors diminishes. In the γ region, solid state Compton detectors can be used to analyze the linear polarization of radiation [12]. Thus, one lacks efficient polarimeters in the region of a few hundred eVs. In the works [7–9], it was proposed to use the process of Rayleigh scattering for the X-ray polarimetry. Therefore, it is important to understand the details of the polarization dependence of the atomic X-ray scattering cross sections. In particular, in [13], it was shown that some remarkable polarization phenomena occur in the process of atomic X-ray scattering, which are absent in the scattering of low energy (e.g., optical) photons. For example, linearly polarized (LP) X-rays, scattered off the closed-shell atoms, could acquire circularly polarized (CP) components. The relative magnitude of the CP component depends on the orientation of the polarization plane of the incident beam, as well as on the scattering angle. Similarly, LP components may occur in the scattering of CP X-rays. These effects could be utilized for the purpose of circular X-ray polarimetry [9]. The mentioned phenomena are manifestations of the circular dichroism (CD) effect, which consists of the dependence of the cross section on the helicity of the photons. Note that for randomly oriented (or spherically symmetric) atomic targets, the CD effects emerge only in differential cross sections, and they disappear after the integration over the photon emission/absorption directions.

The Rayleigh scattering is an example of a two-photon bound–bound process. The polarization properties of such processes were studied in the work [14]. It was shown there

that the CD effects occur not only in the X-ray scattering, but in all two-photon bound–bound transitions including two-photon absorption and decay. For example, CD effects can be observed in the two-photon decay of metastable 2S-states. It is important that such effects emerge only beyond the dipole approximation. Polarization phenomena arising in the process of the two-photon decay of 2S-states of highly charged ions were analyzed in [15]. The reason why various polarization anomalies occur in atomic scattering was discussed in [16,17]. In brief, such effects are caused by the presence of a competing (i.e., "dissipative") channel, such as photoionization. The availability of a competing process results in the emergence of a time-odd parameter (such parameters change sign under the time inversion, $t \to -t$), which leads to the emergence of CD effects. For example, if the energy of the incident photon is higher than the ionization threshold, then the non-dipole effects should be taken into account. This gives rise to the terms in the scattering cross sections, which are responsible for the CD effects. In this case the time-odd parameter is the sine of the relative phase of the scattering amplitudes. (It is represented by the fourth term in the scattering cross section; see Equation (30) below.)

In [14], the polarization dependence of the cross sections was parameterized in terms of the scalar products of complex photon polarization vectors and the unit vectors along the photon momenta, $\hat{\mathbf{k}}_i = \mathbf{k}_i/k_i$, $i = 1, 2$. However, from the experimental point of view, it would be more convenient to have the expressions written in terms of both real vectors defining the photon polarization, as well as the Stokes parameters of light [18,19]. The purpose of this article is twofold. First, we propose the tensor representation for the polarization density matrix (Section 2). The polarization dependence of two-photon processes is described by the product of two-photon density matrices, which is also derived in the tensor form. Second, we applied the derived expression for the product of density matrices for the analysis of the polarization dependence of the X-ray scattering cross section (Section 3). In Section 3.1, we obtain expressions for the polarization parameters of the cross section written in terms of the products of vectors $\hat{\boldsymbol{\epsilon}}_{1,2}$ directed along the major axes of the polarization ellipses of the photons with the unit vectors of photon momenta $\hat{\mathbf{k}}_{1,2}$. In Section 3.2, we write the polarization parameters in terms of the Stokes parameters and discuss the possibility of X-ray polarimetry by means of the atomic scattering process. In the Conclusions (Section 4), we give a brief summary of the derived results and consider their applicability to other problems. Atomic units are used throughout the text.

2. Tensor Representations of the Photon Density Matrix

Below, we derive the tensor form of the photon polarization density matrix, which can be used to re-write the polarization parameters of cross sections for various photoprocesses in terms of scalar products of unit vectors along the photon propagation, $\hat{\mathbf{k}}_{1,2}$, and the directions of the maximal linear polarization, which coincide with the directions of major axes of the photon polarization ellipses in the case of elliptically polarized photons, $\hat{\boldsymbol{\epsilon}}_{1,2}$; see [18].

The polarization state of a photon is often described by a set of three Stokes parameters, ξ_i, so that the photon polarization tensor, $e_\alpha e^*_\beta$ $(\alpha, \beta = x, y)$, in the photon coordinate frame, whose z-axis is directed along the photon propagation $\hat{\mathbf{k}}$, should be replaced by the photon density matrix $\rho_{\alpha\beta}$, which has the form [7,19]:

$$e_\alpha e^*_\beta \to \rho = \frac{1}{2}\begin{pmatrix} 1+\xi_3 & \xi_1 - i\xi_2 & 0 \\ \xi_1 + i\xi_2 & 1-\xi_3 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{1}$$

Here, the photon density matrix is normalized to unity:

$$\mathrm{tr}\ \rho = \sum_{i=1}^{3} e_i e_i^* = (\mathbf{e}\cdot\mathbf{e}^*) = 1. \tag{2}$$

In the theoretical analysis, it is more convenient to use the parameterization of the density matrix in terms of the linear and circular polarization degrees, ℓ and $A = \xi_2$; see [18]. The linear polarization degree is defined by

$$\ell = \sqrt{\xi_1^2 + \xi_3^2}. \tag{3}$$

The Stokes parameters $\xi_{1,3}$ are connected with ℓ by the relations [18]:

$$\xi_1 = \ell \sin 2\varphi, \quad \xi_3 = \ell \cos 2\varphi, \tag{4}$$

where φ is the angle between the x-axis and the direction of the major axis of the photon polarization ellipse, which is described by the unit vector $\hat{\boldsymbol{\epsilon}}$. The components of $\hat{\boldsymbol{\epsilon}}$ are (we consider vectors to be columns composed of their components)

$$\hat{\boldsymbol{\epsilon}} = \begin{pmatrix} \cos\varphi \\ \sin\varphi \\ 0 \end{pmatrix} = (\cos\varphi, \sin\varphi, 0)^\top. \tag{5}$$

In matrix notation, the scalar product of vectors $\mathbf{a}$ and $\mathbf{b}$ has the form $(\mathbf{a} \cdot \mathbf{b}) = \mathbf{a}^\top \mathbf{b}$, so that

$$\hat{\boldsymbol{\epsilon}}^\top \hat{\boldsymbol{\epsilon}} = (\hat{\boldsymbol{\epsilon}} \cdot \hat{\boldsymbol{\epsilon}}) = 1. \tag{6}$$

The photon polarization degree P is defined by [18]

$$P = \sqrt{l^2 + A^2} = \sqrt{\xi_1^2 + \xi_2^2 + \xi_3^2}. \tag{7}$$

For purely polarized light, $P = 1$, while for unpolarized light, $P = 0$. The latter implies that both linear and circular polarization degrees are equal to zero, $\ell = A = 0$.

The symmetric part of the density matrix (1) depends only on the parameter ℓ and the angle φ:

$$\rho_s = \frac{1}{2} \begin{pmatrix} 1 + \ell\cos 2\varphi & \ell \sin 2\varphi & 0 \\ \ell \sin 2\varphi & 1 - \ell \cos 2\varphi & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{8}$$

Let us introduce the unit vector $\hat{\boldsymbol{\epsilon}}'$ by

$$\hat{\boldsymbol{\epsilon}}' = (-\sin\varphi, \cos\varphi, 0)^\top. \tag{9}$$

This vector is directed along the minor axis of the photon polarization ellipse; see Figure 1a. From Equations (5) and (9), we obtain the following useful relations:

$$(\hat{\boldsymbol{\epsilon}}' \cdot \hat{\boldsymbol{\epsilon}}') = 1, \quad (\hat{\boldsymbol{\epsilon}}' \cdot \hat{\boldsymbol{\epsilon}}) = 0, \quad \hat{\boldsymbol{\epsilon}}' = [\hat{\mathbf{k}} \times \hat{\boldsymbol{\epsilon}}]. \tag{10}$$

Now, the density matrix (8) can be written in the covariant form as a sum of two tensors of the second rank:

$$\rho_s = \frac{1+\ell}{2} \hat{\boldsymbol{\epsilon}} \hat{\boldsymbol{\epsilon}}^\top + \frac{1-\ell}{2} \hat{\boldsymbol{\epsilon}}' \hat{\boldsymbol{\epsilon}}'^\top. \tag{11}$$

Our goal is to re-write the second term in (11) in order to avoid the use of the vector $\hat{\boldsymbol{\epsilon}}'$, which is inconvenient in applications. Let $\mathbf{a}$ be an arbitrary vector. Then, the tensor contraction of $a_i a_j$ with the photon density matrix ρ_s is

$$\mathbf{a}^\top \rho_s \mathbf{a} = \sum_{ij} a_i (\rho_s)_{ij} a_j = \frac{1+\ell}{2} (\mathbf{a} \cdot \hat{\boldsymbol{\epsilon}})^2 + \frac{1-\ell}{2} (\mathbf{a} \cdot \hat{\boldsymbol{\epsilon}}')^2. \tag{12}$$

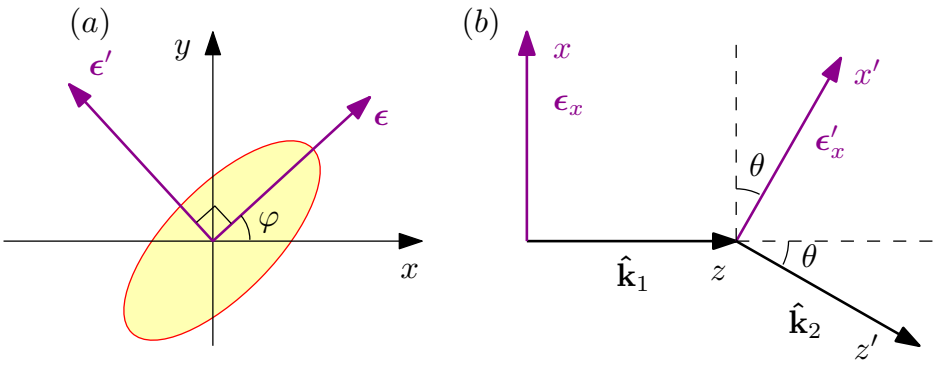


Figure 1. (**a**) Orientation of the unit vectors $\hat{\epsilon}$, $\hat{\epsilon}'$ in the photon polarization plane; the photon propagates towards the reader, $[\hat{\epsilon} \times \hat{\epsilon}'] = \hat{\mathbf{k}}$. (**b**) The scattering geometry. Vectors $\hat{\mathbf{k}}_{1,2}$ define the scattering plane; $0 \leq \theta \leq \pi$ is the scattering angle. (**b**) Coordinate frames of the incident and the scattered photons are shown on the left and right, correspondingly. Unit vectors $\hat{\epsilon}_x$, $\hat{\epsilon}'_x$ define the x-axes of the coordinate frames of the incident and scattered photons; the y-axes of both frames coincide and are directed towards the reader.

Taking into account the coordinate form of the square of a,

$$a^2 = (\mathbf{a} \cdot \hat{\epsilon})^2 + (\mathbf{a} \cdot \hat{\epsilon}')^2 + (\mathbf{a} \cdot \hat{\mathbf{k}})^2, \tag{13}$$

We can exclude the vector $\hat{\epsilon}'$ from the density matrix ρ_s. Namely, by expressing $(\mathbf{a} \cdot \hat{\epsilon}')^2$ from Equation (13) and inserting it into Equation (12), we obtain

$$\mathbf{a}^\top \rho_s \mathbf{a} = \ell\,(\mathbf{a} \cdot \hat{\epsilon})^2 + \ell^{(-)}\left(a^2 - (\mathbf{a} \cdot \hat{\mathbf{k}})^2\right), \quad \ell^{(-)} = \frac{1-\ell}{2}. \tag{14}$$

This equation leads to the following component representation for the symmetric part of the density matrix:

$$(\rho_s)_{ij} = \ell \epsilon_i \epsilon_j + \ell^{(-)}(\delta_{ij} - \hat{k}_i \hat{k}_j). \tag{15}$$

The covariant form of this expression can be written as

$$\rho_s = \ell\, \hat{\epsilon}\hat{\epsilon}^\top + \ell^{(-)}\left(I - \hat{\mathbf{k}}\,\hat{\mathbf{k}}^\top\right). \tag{16}$$

where I is the unit tensor (it is expressed by a *three-by-three* unit matrix). Noting (16), the whole density matrix (1) can be presented in the following tensor form:

$$\rho = \ell\, \hat{\epsilon}\hat{\epsilon}^\top + \ell^{(-)}\left(I - \hat{\mathbf{k}}\,\hat{\mathbf{k}}^\top\right) - \frac{iA}{2}\,[\hat{\mathbf{k}}]_\times, \tag{17}$$

where $[\hat{\mathbf{k}}]_\times$ denotes the skew-symmetric tensor of the second rank, which is defined by the skew-symmetric 3×3 matrix, whose components are given by

$$\left([\hat{\mathbf{k}}]_\times\right)_{ij} = \epsilon_{iqj}\hat{k}_q, \tag{18}$$

where ϵ_{iqj} is the Levi-Civita symbol. The product of the matrix $[\hat{\mathbf{k}}]_\times$ with the column vector $\mathbf{a}$ is equal to the vector cross-product, so that

$$[\hat{\mathbf{k}}]_\times \mathbf{a} = [\hat{\mathbf{k}} \times \mathbf{a}], \quad \mathbf{a}^\top [\hat{\mathbf{k}}]_\times = [\mathbf{a} \times \hat{\mathbf{k}}]^\top. \tag{19}$$

Let $\mathbf{a}$ and $\mathbf{b}$ be arbitrary vectors. Using (19), we can write

$$\mathbf{a}^\top [\hat{\mathbf{k}}_1]_\times\, [\hat{\mathbf{k}}_2]_\times \mathbf{b} = ([\mathbf{a} \times \hat{\mathbf{k}}_1] \cdot [\hat{\mathbf{k}}_2 \times \mathbf{b}]) = (\mathbf{a} \cdot \hat{\mathbf{k}}_2)(\hat{\mathbf{k}}_1 \cdot \mathbf{b}) - (\mathbf{a} \cdot \mathbf{b})(\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2). \tag{20}$$

This equation leads to the following useful relation for the product of two skew-symmetric matrices:

$$[\hat{\mathbf{k}}_1]_\times\,[\hat{\mathbf{k}}_2]_\times = \hat{\mathbf{k}}_2\,\hat{\mathbf{k}}_1^\top - I\cos\theta, \tag{21}$$

where $\cos\theta = (\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)$.

To demonstrate the convenience of the representation (17) of the density matrix, let us analyze the parameter $|\mathbf{e}\cdot\hat{\mathbf{p}}|^2$, where $\hat{\mathbf{p}}$ is a unit vector. For example, this parameter determines the polarization dependence of the photoionization cross section [19]. In this case, $\hat{\mathbf{p}}$ is directed along the photoelectron momentum. We have

$$|\mathbf{e}\cdot\hat{\mathbf{p}}|^2 = (\mathbf{e}\cdot\hat{\mathbf{p}})(\mathbf{e}^*\cdot\hat{\mathbf{p}}) = \hat{\mathbf{p}}^\top\rho_s\hat{\mathbf{p}}. \tag{22}$$

Substituting here the expansion (17) and noting that $(\hat{\mathbf{p}}\cdot[\hat{\mathbf{p}}\times\hat{\mathbf{k}}]) = 0$, we obtain

$$|\mathbf{e}\cdot\hat{\mathbf{p}}|^2 = \ell(\hat{\boldsymbol{\epsilon}}\cdot\hat{\mathbf{p}})^2 + \ell^{(-)}\left(1-(\hat{\mathbf{p}}\cdot\hat{\mathbf{k}})^2\right) = \ell(\hat{\boldsymbol{\epsilon}}\cdot\hat{\mathbf{p}})^2 + \ell^{(-)}[\hat{\mathbf{p}}\times\hat{\mathbf{k}}]^2. \tag{23}$$

In applications, it is often necessary to analyze the parameter $(\mathbf{e}\cdot\mathbf{a})(\mathbf{e}^*\cdot\mathbf{b})$, where $\mathbf{a}$ and $\mathbf{b}$ are vectors entering the problem under consideration, such as the photoelectron momenta vectors etc. The above parameter can be presented as follows:

$$(\mathbf{e}\cdot\mathbf{a})(\mathbf{e}^*\cdot\mathbf{b}) = \mathbf{a}^\top\rho\mathbf{b} = \ell\,(\mathbf{a}\cdot\hat{\boldsymbol{\epsilon}})(\mathbf{b}\cdot\hat{\boldsymbol{\epsilon}}) + \ell^-\,([\mathbf{a}\times\hat{\mathbf{k}}]\cdot[\mathbf{b}\times\hat{\mathbf{k}}]) + \frac{iA}{2}([\mathbf{a}\times\mathbf{b}]\cdot\hat{\mathbf{k}}). \tag{24}$$

In the analysis of two-photon transitions, we have to deal with the product of the density matrices of both photons, $\rho_1\rho_2$. This product can be written in terms of tensor products using the representation (17). Noting the expression (21) for the product of two skew-symmetric matrices, after some transformations, we arrive at the explicit form of the product $\rho_1\rho_2$:

$$\begin{aligned}\rho_1\rho_2 = {}& \ell_1\ell_2\,(\hat{\boldsymbol{\epsilon}}_1\cdot\hat{\boldsymbol{\epsilon}}_2)\,\hat{\boldsymbol{\epsilon}}_1\hat{\boldsymbol{\epsilon}}_2^\top + \ell_1\ell_2^-\,\hat{\boldsymbol{\epsilon}}_1\,[\hat{\mathbf{k}}_2\times[\hat{\boldsymbol{\epsilon}}_1\times\hat{\mathbf{k}}_2]]^\top + \ell_2\ell_1^-\,[\hat{\mathbf{k}}_1\times[\hat{\boldsymbol{\epsilon}}_2\times\hat{\mathbf{k}}_1]]\,\hat{\boldsymbol{\epsilon}}_2^\top \\ & + \ell_1^-\ell_2^-\left(I - \hat{\mathbf{k}}_1\,\hat{\mathbf{k}}_1^\top - \hat{\mathbf{k}}_2\,\hat{\mathbf{k}}_2^\top + \cos\theta\,\hat{\mathbf{k}}_1\,\hat{\mathbf{k}}_2^\top\right) + \frac{A_1A_2}{4}\left(I\cos\theta - \hat{\mathbf{k}}_2\,\hat{\mathbf{k}}_1^\top\right) \\ & + \frac{iA_1}{2}\left(\ell_2[\hat{\mathbf{k}}_1\times\hat{\boldsymbol{\epsilon}}_2]\,\hat{\boldsymbol{\epsilon}}_2^\top - 2\ell_2^-[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]\,\hat{\mathbf{k}}_2^\top + \ell_2^-[\hat{\mathbf{k}}_1]_\times\right) \\ & + \frac{iA_2}{2}\left(\ell_1\hat{\boldsymbol{\epsilon}}_1\,[\hat{\boldsymbol{\epsilon}}_1\times\hat{\mathbf{k}}_2]^\top - 2\ell_1^-\hat{\mathbf{k}}_1\,[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]^\top + \ell_1^-[\hat{\mathbf{k}}_2]_\times\right).\end{aligned} \tag{25}$$

Although this equation is rather lengthy, we emphasize that it describes the polarization dependence of all possible two-photon processes, including light scattering, two-photon single-particle and multiparticle fragmentation, etc. In many particular cases, some of the terms in (25) do not contribute to the final result. For example, the second and third terms in parentheses on the last two lines of (25) do not contribute to the cross sections of two-photon bound–bound transitions in randomly oriented (or spherically symmetric) targets. Indeed, in this case, the cross section contains the polarization parameters $\mathrm{tr}\,(\rho_1\rho_2)$ and $\hat{\mathbf{k}}_\alpha^\top\rho_1\rho_2\hat{\mathbf{k}}_\beta$, $\alpha,\beta = 1,2$. These parameters involving the second and the third terms in the last line of (25) have the form:

$$\begin{aligned}\mathrm{tr}\,(\hat{\mathbf{k}}_1\,[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]^\top) &= (\hat{\mathbf{k}}_1\cdot[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]) = 0, \\ \hat{\mathbf{k}}_\alpha^\top\,\hat{\mathbf{k}}_1\,[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]^\top\,\hat{\mathbf{k}}_\beta &= ([\hat{\mathbf{k}}_\alpha\times\hat{\mathbf{k}}_1]\cdot[[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]\times\hat{\mathbf{k}}_\beta]) = 0, \\ \hat{\mathbf{k}}_\alpha^\top\,[\hat{\mathbf{k}}_2]\,\hat{\mathbf{k}}_\beta &= (\hat{\mathbf{k}}_\alpha\cdot[\hat{\mathbf{k}}_2\times\hat{\mathbf{k}}_\beta]) = 0, \quad \alpha,\beta = 1,2.\end{aligned} \tag{26}$$

The similar terms in the third line of (25) are also equal to zero. The reason for those terms vanishing is geometrical. The cross section is a true scalar quantity, while the circular polarization degree A is a pseudoscalar [19]. Thus, it can enter the cross section only as a product with another pseudoscalar, which can only be composed out of three (or more) differently oriented vectors. In the case considered, there are only *two* such vectors: $\hat{\mathbf{k}}_1$

and $\hat{\mathbf{k}}_2$. However, for bound-free transitions, such as ionization or recombination, there is an additional vector $\hat{\mathbf{p}} = \mathbf{p}/p$, where $\mathbf{p}$ is the free electron's momentum vector, and the additional pseudoscalar quantity is $(\hat{\mathbf{p}} \cdot [\hat{\mathbf{k}}_1 \times \hat{\mathbf{k}}_2])$. Therefore, for bound-free non-dipole transitions including two different partially polarized photons, all terms in Equation (25) contribute to the cross section.

For both LP photons $\ell_1 = \ell_2 = 1$, $A_1 = A_2 = 0$, and the product of the density matrices (25) simplifies to

$$\rho_1^{(LP)} \rho_2^{(LP)} = (\hat{\boldsymbol{\epsilon}}_1 \cdot \hat{\boldsymbol{\epsilon}}_2)\, \hat{\boldsymbol{\epsilon}}_1 \hat{\boldsymbol{\epsilon}}_2^{\top}. \tag{27}$$

For both CP photons $\ell_1 = \ell_2 = 0$, $A_{1,2} = \pm 1$, and (25) becomes

$$\begin{aligned} 4\rho_1^{(CP)} \rho_2^{(CP)} = I - \hat{\mathbf{k}}_1 \hat{\mathbf{k}}_1^{\top} - \hat{\mathbf{k}}_2 \hat{\mathbf{k}}_2^{\top} + \cos\theta\, \hat{\mathbf{k}}_1 \hat{\mathbf{k}}_2^{\top} + A_1 A_2 \Big(I \cos\theta - \hat{\mathbf{k}}_2 \hat{\mathbf{k}}_1^{\top} \Big) \\ + i A_1 \Big([\hat{\mathbf{k}}_1]_{\times} - 2[\hat{\mathbf{k}}_1 \times \hat{\mathbf{k}}_2] \hat{\mathbf{k}}_2^{\top} \Big) + i A_2 \Big([\hat{\mathbf{k}}_2]_{\times} - 2\hat{\mathbf{k}}_1 [\hat{\mathbf{k}}_1 \times \hat{\mathbf{k}}_2]^{\top} \Big). \end{aligned} \tag{28}$$

For unpolarized photons $\ell_{1,2} = A_{1,2} = 0$, and the last three terms in the above equation vanish.

3. Polarization Dependence of the Cross Section of X-ray Scattering

For simplicity, we restrict our consideration only to the case of initial and final S-states, which is the most important in the process of the scattering of X- and γ-rays by atoms [2,20].

For transitions between the states having zero total angular momentum, the scattering amplitude can be parameterized as follows [14]:

$$A = f_1(\theta)\, (\mathbf{e}_1 \cdot \mathbf{e}_2^*) + f_2(\theta)\, (\mathbf{e}_1 \cdot \hat{\mathbf{k}}_2)(\mathbf{e}_2^* \cdot \hat{\mathbf{k}}_1), \tag{29}$$

where both functions, $f_1(\theta)$ and $f_2(\theta)$, depend on the angle θ between the unit vectors $\hat{\mathbf{k}}_1$ and $\hat{\mathbf{k}}_2$ defining the directions of the incident ($\mathbf{e}_1$) and the scattered ($\mathbf{e}_2$) photon beams. Note that for the two-photon decay process, one has to replace $\mathbf{e}_1 \to \mathbf{e}_1^*$, while for the two-photon absorption process, the substitution $\mathbf{e}_2^* \to \mathbf{e}_2$ should be made in Equation (29). The scattering cross section corresponding to the amplitude (29) can be written in the form [14]:

$$\frac{d\sigma}{d\Omega_{\hat{\mathbf{k}}_2}} = r_0^2 \frac{\omega_2}{\omega_1} \Big[\mathcal{P}_1 |f_1|^2 + \mathcal{P}_2 |f_2|^2 + 2\mathcal{P}_3\, \mathrm{Re}\,(f_1 f_2^*) - 2\mathcal{P}_3'\, \mathrm{Im}\,(f_1 f_2^*) \Big], \tag{30}$$

where r_0 is the classical electron radius, ω_1 (ω_2) is the frequency of the incident (scattered) photon (for elastic, i.e., Rayleigh, scattering, $\omega_2 = \omega_1$), and the $\mathcal{P}$'s are the polarization parameters defined by

$$\begin{aligned} \mathcal{P}_1 &= |\mathbf{e}_1 \cdot \mathbf{e}_2^*|^2, \\ \mathcal{P}_2 &= |\mathbf{e}_1 \cdot \hat{\mathbf{k}}_2|^2\, |\mathbf{e}_2^* \cdot \hat{\mathbf{k}}_1|^2, \\ \mathcal{P}_3 &= \mathrm{Re}\,\{(\mathbf{e}_1 \cdot \mathbf{e}_2^*)(\mathbf{e}_1^* \cdot \hat{\mathbf{k}}_2)(\mathbf{e}_2 \cdot \hat{\mathbf{k}}_1)\}, \\ \mathcal{P}_3' &= \mathrm{Im}\,\{(\mathbf{e}_1 \cdot \mathbf{e}_2^*)(\mathbf{e}_1^* \cdot \hat{\mathbf{k}}_2)(\mathbf{e}_2 \cdot \hat{\mathbf{k}}_1)\}. \end{aligned} \tag{31}$$

Equation (30) completely describes the dependence of the cross section on the polarization states of the photons. Note that in the electric dipole approximation only, the first term on the r.h.s. of Equation (30) is non-zero.

3.1. Cross Section in the Vector Form

Below, we present the polarization coefficients of the cross section in the vector form as combinations of scalar products of real unit vectors along the directions of the photon propagation and the major linear polarization. The first term on the right-hand side of

(30) is a trace of the matrix product (25). In this case, the terms in the last two lines of Equation (25) cancel out, and we obtain

$$\mathcal{P}_1 = \mathrm{tr}\,(\rho_1\rho_2) = \ell_1\ell_2(\hat{\boldsymbol{e}}_1\cdot\hat{\boldsymbol{e}}_2)^2 + \ell_1\ell_2^-[\hat{\boldsymbol{e}}_1\times\hat{\mathbf{k}}_2]^2 + \ell_2\ell_1^-[\hat{\boldsymbol{e}}_2\times\hat{\mathbf{k}}_1]^2 + \ell_1^-\ell_2^-\left(1+\cos^2\theta\right) + \frac{A_1A_2}{2}\cos\theta. \quad (32)$$

After some elementary transformations, this equation can be re-written in the following explicit form:

$$\mathcal{P}_1 = \frac{1-\ell_1\ell_2}{2} + \ell_1\ell_2(\hat{\boldsymbol{e}}_1\cdot\hat{\boldsymbol{e}}_2)^2 - \frac{\ell_1(1-\ell_2)}{2}(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)^2 - \frac{\ell_2(1-\ell_1)}{2}(\hat{\boldsymbol{e}}_2\cdot\hat{\mathbf{k}}_1)^2 - \frac{(1-\ell_1)(1-\ell_2)}{4}\sin^2\theta + \frac{A_1A_2}{2}\cos\theta. \quad (33)$$

We remind that, in the dipole approximation, the only non-zero parameter in the cross section (30) is $\mathcal{P}_1$. For purely linearly polarized photons, we have $\ell_1 = \ell_2 = 1$, and (33) reduces to

$$|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2_{\mathrm{LP}} = (\hat{\boldsymbol{e}}_1\cdot\hat{\boldsymbol{e}}_2)^2. \quad (34)$$

For unpolarized or purely circularly polarized photons, we have $\ell_1 = \ell_2 = 0$ and

$$|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2_{\mathrm{CP}} = \frac{\cos^2\theta+1}{2} + \frac{A_1A_2}{2}\cos\theta. \quad (35)$$

The second polarization parameter in (30) can be expressed using Equation (23):

$$\mathcal{P}_2 = \left(\ell_1(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)^2 + \frac{1-\ell_1}{2}\sin^2\theta\right)\left(\ell_2(\hat{\boldsymbol{e}}_2\cdot\hat{\mathbf{k}}_1)^2 + \frac{1-\ell_2}{2}\sin^2\theta\right). \quad (36)$$

Now, we turn to the analysis of the interference coefficient, which determines the last two terms in the cross section (30). First, we re-write it via the product of density matrices $\rho_1\rho_2$:

$$\mathcal{P}_3 = 2\left(\hat{\mathbf{k}}_2^\top\rho_1\rho_2\hat{\mathbf{k}}_1\right)^*. \quad (37)$$

Next, we employ the expression (25) for the product of density matrices $\rho_1\rho_2$, which yields

$$\begin{aligned}\mathcal{P}_3 =& \ell_1\ell_2(\hat{\boldsymbol{e}}_1\cdot\hat{\boldsymbol{e}}_2)\,(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)\,(\hat{\boldsymbol{e}}_2\cdot\hat{\mathbf{k}}_1) - \ell_1\ell_2^-(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)^2\cos\theta - \ell_2\ell_1^-(\hat{\boldsymbol{e}}_2\cdot\hat{\mathbf{k}}_1)^2\cos\theta \\ & - \ell_1^-\ell_2^-\cos\theta\sin^2\theta - \frac{A_1A_2}{4}\sin^2\theta, \\ \mathcal{P}_3' =& \frac{A_1\ell_2}{2}(\hat{\boldsymbol{e}}_2\cdot\hat{\mathbf{k}}_1)\,(\hat{\boldsymbol{e}}_2\cdot[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]) + \frac{A_2\ell_1}{2}(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)\,(\hat{\boldsymbol{e}}_1\cdot[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]).\end{aligned} \quad (38)$$

It is interesting to analyze the case of a linearly polarized incident photon (LP1), which corresponds to $\ell_1 = 1$, $A_1 = 0$. In this case, the polarization parameters (33), (36) and (38) reduce to

$$\begin{aligned}\mathcal{P}_1^{(LP1)} &= \frac{1-\ell_2}{2}[\hat{\boldsymbol{e}}_1\times\hat{\mathbf{k}}_2]^2, \\ \mathcal{P}_2^{(LP1)} &= (\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)^2\left(\ell_2(\hat{\boldsymbol{e}}_2\cdot\hat{\mathbf{k}}_1)^2 + \frac{1-\ell_2}{2}\sin^2\theta\right), \\ \mathcal{P}_3^{(LP1)} &= \ell_2(\hat{\boldsymbol{e}}_1\cdot\hat{\boldsymbol{e}}_2)\,(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)\,(\hat{\boldsymbol{e}}_2\cdot\hat{\mathbf{k}}_1) + \frac{1-\ell_2}{2}(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)^2\cos\theta, \\ \mathcal{P}_3'^{(LP1)} &= \frac{A_2}{2}(\hat{\boldsymbol{e}}_1\cdot\hat{\mathbf{k}}_2)\,(\hat{\boldsymbol{e}}_1\cdot[\hat{\mathbf{k}}_1\times\hat{\mathbf{k}}_2]).\end{aligned} \quad (39)$$

The mean value of the circular polarization degree of the scattered light is defined by the equation:

$$\langle A_2 \rangle = \frac{I(A_2 = +1) - I(A_2 = -1)}{I(A_2 = +1) + I(A_2 = -1)}, \tag{40}$$

where $I(A_2) \sim d\sigma/d\Omega_{\hat{\mathbf{k}}_2}$ is the intensity of light scattered in the direction $\hat{\mathbf{k}}_2$. Noting Equations (30) and (39), we can write

$$\langle A_2 \rangle = C_A(\theta)\,(\hat{\boldsymbol{\epsilon}}_1 \cdot \hat{\mathbf{k}}_2)\,(\hat{\boldsymbol{\epsilon}}_1 \cdot [\hat{\mathbf{k}}_1 \times \hat{\mathbf{k}}_2]), \tag{41}$$

where the coefficient C_A is independent of the polarization of photons, and it is defined by

$$C_A(\theta) = \frac{2\,\mathrm{Im}\,(f_1 f_2^*)}{[\hat{\boldsymbol{\epsilon}}_1 \times \hat{\mathbf{k}}_2]^2\,|f_1|^2 + (\hat{\boldsymbol{\epsilon}}_1 \cdot \hat{\mathbf{k}}_2)^2\,(\sin^2\theta\,|f_2|^2 + 2\cos\theta\ \mathrm{Re}\,(f_1 f_2^*))}. \tag{42}$$

From (41) and (42), it is seen that the linearly polarized light, scattered off the randomly oriented atomic target, acquires a chiral component whose magnitude depends on the scattering angle and the orientation of the polarization plane of the incident light. From (40), it follows that this CD effect is absent in the long wave (i.e., dipole) approximation. The properties of the vector dot products in (41) lead to the conclusion that the CD is missing when the incident light is polarized in the scattering plane or perpendicularly to it. It is remarkable that the helicity (i.e., the sign of the chiral component, sign $\langle A_2 \rangle$) of scattered light depends not only on the geometric factor in (41), but also on the relative phase of the complex amplitude parameters, $\arg(f_1 f_2^*)$, which depends on the scattering angle θ.

3.2. Cross Section in Terms of Stokes Parameters

From the experimental point of view, it is often more convenient to have expressions for the polarization parameters (33), (36) and (38) written in terms of Stokes parameters. In order to obtain such expressions, it is necessary to specify the x- and y-axes of the coordinate frames connected with the polarization planes of the incident and scattered photons. Our choice of these coordinate frames is shown in Figure 1b. Namely, the z-axes are defined by the photon momenta vectors $\mathbf{k}_1$ and $\mathbf{k}_2$; both y-axes coincide, and they are perpendicular to the scattering plane; the x- and x'-axes lie in the scattering plane, so that their mutual angle is equal to the scattering angle θ. As was shown above, the polarization parameters are defined by equations involving the product of photon density matrices, $\rho_1\rho_2$. However, we cannot directly use their representation in terms of Stokes parameters (1), since it is valid only in a particular coordinate frame whose z-axis is directed along the photon momentum $\mathbf{k}$. Below, we calculate the polarization parameters in the coordinate frame of the scattered photon; see Figure 1b. In that frame, the density matrix of the second (i.e., scattered) photon is given by (1), while the density matrix of the first (i.e., incident) photon is defined by

$$\rho_1 = R\,\rho(\xi^{(1)})\,R^\top, \tag{43}$$

where $\rho(\xi^{(1)})$ is given by (1) (with the obvious replacements $\xi_i \to \xi_i^{(1)}$) and R is the rotation matrix corresponding to the rotation from the coordinate frame of the incident photon to that of the scattered one. From Figure 1b, it is seen that the Euler angles describing this rotation are $(\alpha, \beta, \gamma) = (0, \theta, 0)$. Thus, the product $\rho_1\rho_2$ should be calculated according to the formula:

$$\rho_1\rho_2 = R(\theta)\,\rho(\xi^{(1)})\,R^\top(\theta)\,\rho(\xi^{(2)}), \tag{44}$$

where the arguments $\xi^{(1)}$, $\xi^{(2)}$ indicate that the corresponding density matrix should be taken in the form of (1). The components of the vectors $\hat{\mathbf{k}}_{1,2}$ in the coordinate frame of the scattered photon are

$$\hat{\mathbf{k}}_1 = (\sin\theta, 0, \cos\theta)^\top, \quad \hat{\mathbf{k}}_2 = (0, 0, 1)^\top. \tag{45}$$

Now, we are able to express the polarization parameters (32), (36), and (37) via the Stokes parameters by calculating the corresponding matrix products, which can be performed using any modern computer algebra software. Below, we present the final results of the computations. Namely, the first (i.e., electric dipole) polarization parameter is

$$\mathcal{P}_1 = \frac{\cos\theta}{2}\left(\xi_1^{(1)}\xi_1^{(2)} + \xi_2^{(1)}\xi_2^{(2)}\right) + \frac{\cos^2\theta}{4}\,(1+\xi_3^{(1)})(1+\xi_3^{(2)}) + \frac{(1-\xi_3^{(1)})(1-\xi_3^{(2)})}{4}. \quad (46)$$

The second polarization parameter has the form

$$\mathcal{P}_2 = \frac{\sin^4\theta}{4}(1+\xi_3^{(1)})(1+\xi_3^{(2)}). \quad (47)$$

The interference terms are

$$\mathcal{P}_3 = -\frac{\sin^2\theta}{4}\left(\xi_1^{(1)}\xi_1^{(2)} + \xi_2^{(1)}\xi_2^{(2)} + (1+\xi_3^{(1)})(1+\xi_3^{(2)})\,\cos\theta\right), \quad (48)$$

$$\mathcal{P}_3' = \frac{\sin^2\theta}{4}\left(\xi_2^{(1)}\xi_1^{(2)} - \xi_1^{(1)}\xi_2^{(2)}\right). \quad (49)$$

The expression for the scattering cross section takes a rather compact form when written in terms of the "parallel" and "perpendicular" amplitude parameters [2,21]:

$$A_\perp = f_1, \quad A_\parallel = f_1\cos\theta - f_2\,\sin^2\theta. \quad (50)$$

Inserting into Equation (30) the parameters (46)–(49) and noting the definitions (50), the cross section becomes [2,21]:

$$\frac{d\sigma}{d\Omega_{\mathbf{k}_2}} = \frac{r_0^2}{4}\frac{\omega_2}{\omega_1}\Big[(1-\xi_3^{(1)})(1-\xi_3^{(2)})\,|A_\perp|^2 + (1+\xi_3^{(1)})(1+\xi_3^{(2)})\,|A_\parallel|^2 + 2\left(\xi_1^{(1)}\xi_1^{(2)} + \xi_2^{(1)}\xi_2^{(2)}\right)\mathrm{Re}\,(A_\perp A_\parallel^*) + 2\left(\xi_2^{(1)}\xi_1^{(2)} - \xi_1^{(1)}\xi_2^{(2)}\right)\mathrm{Im}\,(A_\perp A_\parallel^*)\Big]. \quad (51)$$

We emphasize that this equation is valid only for the geometry defined by Figure 1b. This is in contrast to Equations (33), (36) and (38), which are independent of the choice of the coordinate frames.

Let us consider the CD effect when the incident light is purely linearly polarized, $\xi_2^{(1)} = 0$. As was already mentioned above, the CD vanishes if the polarization plane of the incident light coincides, or is perpendicular, to the scattering plane. In our geometry (see Figure 1b), these situations correspond to $\xi_3^{(1)} = \pm 1$, and we exclude them from consideration. Next, we assume that only circularly polarized photons are detected, which means $\xi_1^{(2)} = \xi_3^{(2)} = 0$ and $\xi_2^{(2)} = \pm 1$. Under these conditions, Equation (30) for the cross section reduces to

$$\left.\frac{d\sigma}{d\Omega_{\mathbf{k}_2}}\right|_{\xi_2^{(2)}=\pm 1} = \frac{r_0^2}{4}\frac{\omega_2}{\omega_1}\left[S_3 - 2\xi_1^{(1)}\xi_2^{(2)}\,\mathrm{Im}\,(A_\perp A_\parallel^*)\right], \quad S_3 \equiv (1-\xi_3^{(1)})\,|A_\perp|^2 + (1+\xi_3^{(1)})\,|A_\parallel|^2. \quad (52)$$

The circular polarization degree of the scattered light (or the relative CD), defined by (40), takes the form

$$\langle \xi_2^{(2)} \rangle \equiv \langle A_2 \rangle = \frac{I_{\xi_2^{(2)}=+1} - I_{\xi_2^{(2)}=-1}}{I_{\xi_2^{(2)}=+1} + I_{\xi_2^{(2)}=-1}} = -2\xi_1^{(1)} \frac{\mathrm{Im}\,(A_\perp A_\parallel^*)}{S_3}. \tag{53}$$

According to Equation (4), $\xi_1^{(1)} = \pm 1$ means that the polarization plane of the incident light is tilted at an angle $\pm\pi/4$ with respect to the scattering plane. Thus, the helicity of the scattered light is determined by the position of the polarization plane of the incident light. This effect does not exist in the dipole approximation, since the second amplitude parameter is zero in this case, $f_2 = 0$.

In the recent work [9], it was proposed to use Rayleigh scattering to measure the degree of the circular polarization of X-rays. In [9], the analysis was performed under the assumption that the incident light is completely circularly polarized. Equations (46)–(48) allow answering the general question on how to determine the polarization parameters of the incident light when only linearly polarized scattered light is detected. If the scattered light is polarized in the scattering plane or perpendicularly to it, then $\xi_3^{(2)} = \pm 1$, $\xi_2^{(2)} = \xi_1^{(2)} = 0$, and the cross section has the form

$$\left.\frac{d\sigma}{d\Omega_{\hat{\mathbf{k}}_2}}\right|_{\xi_3^{(2)}=\pm 1} = \frac{r_0^2}{4}\frac{\omega_2}{\omega_1}\left[(1-\xi_3^{(1)})(1-\xi_3^{(2)})\,|A_\perp|^2 + (1+\xi_3^{(1)})(1+\xi_3^{(2)})\,|A_\parallel|^2\right]. \tag{54}$$

It is seen that, by taking the ratio of two relative values of the scattered light intensity, corresponding to $\xi_3^{(2)} = \pm 1$, one can determine the value of $\xi_3^{(1)}$:

$$\langle \xi_3^{(2)} \rangle = \frac{I_{\xi_3^{(2)}=+1} - I_{\xi_3^{(2)}=-1}}{I_{\xi_3^{(2)}=+1} + I_{\xi_3^{(2)}=-1}} = \frac{1}{S_3}\left[(1+\xi_3^{(1)})|A_\parallel|^2 - (1-\xi_3^{(1)})\,|A_\perp|^2\right]. \tag{55}$$

According to (54), $\xi_3^{(1)}$ could be evaluated knowing only one atomic parameter, $|A_\perp|/|A_\parallel|$. To determine two remaining parameters, $\xi_1^{(1)}$ and $\xi_2^{(1)}$, one has to measure the intensity of the scattered light, polarized in planes tilted at the angles $\pm\pi/4$ with respect to the scattering plane. This means that $\xi_1^{(2)} = \pm 1$, $\xi_2^{(2)} = \xi_3^{(2)} = 0$, and the corresponding cross section is

$$\left.\frac{d\sigma}{d\Omega_{\hat{\mathbf{k}}_2}}\right|_{\xi_1^{(2)}=\pm 1} = \frac{r_0^2}{4}\frac{\omega_2}{\omega_1}\left[S_3 + 2\xi_1^{(1)}\xi_1^{(2)}\,\mathrm{Re}\,(A_\perp A_\parallel^*) + 2\xi_2^{(1)}\xi_1^{(2)}\,\mathrm{Im}\,(A_\perp A_\parallel^*)\right]. \tag{56}$$

It is clear that the values of the two parameters $\xi_1^{(1)}$, $\xi_2^{(1)}$ cannot be extracted from a single value of the relative intensity, which is

$$\langle \xi_1^{(2)} \rangle = \frac{I_{\xi_1^{(2)}=+1} - I_{\xi_1^{(2)}=-1}}{I_{\xi_1^{(2)}=+1} + I_{\xi_1^{(2)}=-1}} = \frac{2}{S_3}\left[\xi_1^{(1)}\,\mathrm{Re}\,(A_\perp A_\parallel^*) + \xi_2^{(1)}\,\mathrm{Im}\,(A_\perp A_\parallel^*)\right]. \tag{57}$$

Thus, one has to perform several measurements of $\langle \xi_1^{(2)} \rangle$ for different values of the scattering angle θ. This implies that the values of the amplitude parameters $|A_\perp|/|A_\parallel|$ and $\arg(A_\perp A_\parallel^*)$ should be known for those angles. The above expressions for the Stokes parameters are valid only for the scattering by closed shells. The scattering amplitude for open shells differs from that given by Equation (30), so that the cross section contains not four (as in (30)), but eight different polarization parameters [14]. These additional parameters can be expressed similarly to (33)–(38) using Equation (25) for the product of density matrices. Alternatively, they can be written in terms of Stokes parameters by calculating the matrix product given by Equation (44).

4. Conclusions

We analyzed the polarization dependence of two-photon transitions using the tensor representation of the photon polarization density matrix given by Equation (17). This representation makes it possible to write the polarization parameters of the cross sections in terms of the scalar products of the unit vectors defining the directions of the photon propagation $\hat{\mathbf{k}}_{1,2}$ and the major axes of the photon polarization ellipses, $\hat{\boldsymbol{\epsilon}}_{1,2}$. The advantage of vector representations of polarization parameters, similar to those given by Equations (33)–(38), is that they simplify the analysis of various specific cases needed in applications. For example, for unpolarized or circularly polarized photons, all terms with vectors $\hat{\boldsymbol{\epsilon}}_{1,2}$ cancel out, and only terms with $\hat{\mathbf{k}}_{1,2}$ enter the cross section, etc.

The polarization dependence of the two-photon processes is determined by the product of two density matrices, $\rho_1\rho_2$. For example, in the case of two-photon one-electron ionization, the polarization dependence is determined by the matrix product $\hat{\mathbf{p}}^\top\rho_1\rho_2\hat{\mathbf{p}}$, where $\hat{\mathbf{p}}$ is the column with the coordinates of the unit vector along the photoelectron momentum. The tensor form of the product $\rho_1\rho_2$, given by Equation (25), allows one to write such matrix products in terms of vector dot products involving $\hat{\mathbf{k}}$, $\hat{\mathbf{e}}$ and other vectors entering the problem (as $\hat{\mathbf{p}}$ in the above example). We note that the polarization dependence of two-photon processes could be rather complicated, which is reflected by a rather lengthy form of Equation (25). Namely, the cross section of the X-ray scattering on the open-shell atoms involves *eight* different polarization parameters [14]. Four of them are the parameters $\mathcal{P}_1$–$\mathcal{P}_3$, $\mathcal{P}'_3$, given by (33)–(38), and the four others can be obtained from $\mathcal{P}_1$–$\mathcal{P}_3$, $\mathcal{P}'_3$ by the interchange $\mathbf{e}_{1,2} \leftrightarrow \mathbf{e}^*_{1,2}$. This interchange means that the replacements $A_{1,2} \to -A_{1,2}$ should be made in Equations (33)–(38). A similarly rich polarization structure has the cross section of the two-photon two-electron ionization of atoms [22–27].

Often, it is convenient to parameterize the polarization factors in terms of the Stokes parameters of the two photons, $\xi_i^{(1)}$, $\xi_i^{(2)}$, $i = 1, 2, 3$. Accordingly, it is necessary to have the product of $\rho_1\rho_2$ written in terms of $\xi_i^{(1)}$, $\xi_i^{(2)}$. For two different photons (as in the scattering process), the product $\rho_1\rho_2$ cannot be calculated directly by using the definition of the density matrix (1), since it is only valid in the coordinate frame with the z-axis directed along the photon propagation. This problem was addressed in Section 3.2, where we described the procedure for the calculation of the product $\rho_1\rho_2$. This procedure is also applicable to processes with a larger number of photons. Namely, the product $\rho_1\rho_2\rho_3$, which determines the polarization factors in three-photon processes, could be calculated by inserting rotation matrices, describing rotations from the photon to the laboratory coordinate frames, similarly to Equation (44).

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Abbreviations

The following abbreviations are used in this manuscript:

CD Circular dichroism
CP Circular polarization
LP Linear polarization

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