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OPTICS AND LASER PHYSICS

Two-Photon Ionization of Single Atoms Localized on the Bessel Beam Axis

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In the article the effect of twisting of Bessel radiation on two-photon ionization of single atoms localized on the axis of the incident beam is studied. The matrix element of two-photon ionization of this type is obtained for arbitrary polarization and multipolarity of the incident radiation. The differential and integral probability of ionization of an atom over the photoemission angle is analyzed. Illustrative calculations are performed for helium and neon atoms in the simplest case of a circularly polarized field in the electric dipole approximation in the single-active-electron model.

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1. INTRODUCTION

Photoelectron emission studies—a powerful and well-developed method for studying the nature and internal structure of matter—have reached a qualitatively new level with the development of methods for detecting vector correlations, such as angular distributions of reaction products, electron spin polarization, or various types of dichroism [1]. The spectrum of possible phenomena becomes even broader when it comes to multiphoton processes, since the absorption of the first photon creates a polarized target with a certain non-uniform distribution of electron density in space, and subsequent photons act as a probe for the primary absorption [2–4]. Until recently, the main factor influencing the kinematics of multiphoton processes was the polarization of radiation [5–10]. However, with the development of methods for generating twisted radiation, new prospects have opened up [11–15]. Twisted radiation has unique characteristics such as a non-uniform radiation intensity profile, a non-plane constant phase surface, a complex structure of energy flows inside the beam and one can expect that these characteristics will manifest themselves in a special way in the angular distributions of photoemission. To date, the literature on studies of two-photon processes involving twisted radiation is represented by a limited number of works and mainly relates to a combination of twisted light and a plane wave [16–18].

For the analysis, we selected photoionization by Bessel light propagating along the z axis (quantization axis) with the atom localized on the same axis. Such geometry was considered, e.g., in [19–22]. In this case, the Bessel state of radiation is characterized by a linear projection of the momentum k_z and a projection of the total angular momentum $m_{\rm tam}$ onto the z axis. The absolute value of the transverse momentum, $\varkappa_\perp = |\mathbf{k}_\perp|$, is fixed and, together with k_z, determines the energy of the incident radiation: $\omega = c\sqrt{\varkappa_\perp^2 + k_z^2}$, where c is the speed of light in vacuum. As shown in [22], such a Bessel state is described by the vector potential

$$\mathbf{A}^{\rm tw}_{\varkappa_\perp m_{\rm tam}\lambda} = \int \mathbf{u}_\lambda e^{i\mathbf{kr}} a_{\varkappa_\perp m_{\rm tam}}(\mathbf{k}_\perp)\frac{d^2\mathbf{k}_\perp}{4\pi^2}, \qquad (1)$$

where

$$a_{\varkappa_\perp m_{\rm tam}}(\mathbf{k}_\perp) = (-i)^{m_{\rm tam}} e^{i m_{\rm tam}\phi_k}\sqrt{\frac{2\pi}{k_\perp}}\delta(k_\perp - \varkappa_\perp), \qquad (2)$$

and $\mathbf{u}_\lambda$ is the polarization vector with helicity $\lambda = \pm 1$.

The Bessel wave defined by Eqs. (1) and (2) in momentum space can be represented as a coherent superposition of plane waves whose wave vectors $\mathbf{k} = (\mathbf{k}_\perp, k_z)$ lie on the surface of a certain cone with an opening angle $\tan\theta_c = k_\perp/k_z$ (see Fig. 1). The position of the irradiated target in the xy plane can be specified by the vector $\mathbf{b}_\perp$.

2. RESULTS

In this paper, we obtain a general expression that allows us to describe the angular distribution of photoelectrons produced in two-photon ionization of an

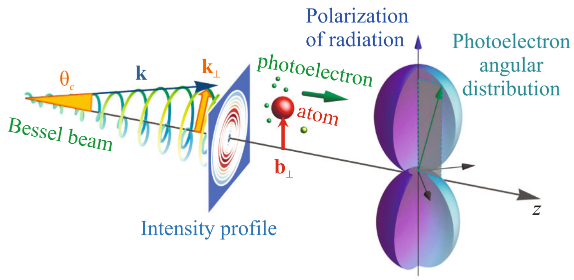

Fig. 1. (Color online) General scheme of ionization of an atom by Bessel radiation.

atom by Bessel waves, when the atom is located on the axis of the incident beam (see Eq. (A.9) in the Appendix). By the general expression we mean that the electromagnetic interaction of arbitrary multipolarity and polarization is considered. Here, for illustration, we consider ionization by a circularly polarized field in the dipole approximation and single active electron approximation (details of the derivation, as well as the notation used, are given in the Appendix):

$$\begin{aligned}\widetilde{W}^{(\mathrm{tw,circ})}_{2\gamma,E1}(\theta_p;\theta_c) &= 45\times\sum_{\substack{L_\gamma L'_\gamma k_\gamma r\\ l_\xi l'_\xi l l'}}(-1)^{L_f+l+l'}\frac{\hat{l}\hat{l}'}{\hat{L}_f^2}\\ &\times(11,1-1\,|\,L_\gamma 0)^2(11,1-1\,|\,L'_\gamma 0)^2(L_\gamma 0,L'_\gamma 0\,|\,k_\gamma 0)^2\\ &\times(22,2-2\,|\,r0)(l0,l'0\,|\,r0)\begin{Bmatrix}L_f & l & 2\\ 1 & 1 & l_\xi\end{Bmatrix}\begin{Bmatrix}L_f & l' & 2\\ 1 & 1 & l'_\xi\end{Bmatrix}\\ &\times\begin{Bmatrix}l & l' & r\\ 2 & 2 & L_f\end{Bmatrix}\times P_r(\cos\theta_p)P_{k_\gamma}(\cos\theta_c)\mathcal{R}_{l_0 l_\xi l}\mathcal{R}^*_{l_0 l'_\xi l'},\end{aligned}\qquad(3)$$

where $P_n(x)$ is Legendre polynomial of the order n, $\mathcal{R}_{l_0 l_\xi l}$ is the radial integral of the corresponding matrix element.

The two-photon ionization amplitudes were calculated using the method proposed in [23, 24]. This method is also described in detail in [25], however, it is worth noting that the summation in Eq. (5) of this paper must be started with $m = 0$. The wavefunctions were calculated in the RADIAL program [26] using potential of $2p$-electron in $1s2p$ (for helium) and $1s^2 2s^2 2p^5 3p$ (for neon) configurations, obtained within the MCHF software package [27].

Ionization of the 1*s Shell of Helium Atom*

According to Eq. (3), the photoelectron angular distribution during ionization of the $1s$ shell of a helium atom by two Bessel waves will have the form

$$\widetilde{W}^{(tw)}_{2\gamma,E1}(\theta_p;\theta_c) = \frac{9}{8}\cos^8\frac{\theta_c}{2}\sin^4\theta_p|\mathcal{R}_{spd}|^2,\qquad(4)$$

where $\mathcal{R}_{spd}$ corresponds to $\mathcal{R}_{l_0 l_\xi l}$ with $l_0 = 0$; $l_\xi = 1$ and $l = 2$.

The cross section for the process in this case:

$$\tilde{\sigma}^{(\mathrm{tw})}_{2\gamma,E1}(\theta_c) = \frac{3}{5}\cos^8\frac{\theta_c}{2}|\mathcal{R}_{spd}|^2.\qquad(5)$$

Note that in the case of double ionization of the s-shell by a circularly polarized field, the only allowed channel is the εd wave. The cross sections and photoelectron angular distributions corresponding to Eqs. (4) and (5) are shown in Fig. 2.

Ionization of the Neon Atom 2*p Shell*

According to Eq. (3), the photoelectron angular distribution during ionization of the $2p$ shell of a neon atom ($l_0 = 1$) by two Bessel waves will have the form

$$\begin{aligned}\widetilde{W}^{(\mathrm{tw})}_{2\gamma,E1}(\theta_p;\theta_c) &= \frac{1}{800}\cos^8\frac{\theta_c}{2}\sin^2\theta_p\\ &\times\Big(400|\mathcal{R}_{psp}|^2 + 4|\mathcal{R}_{pdp}|^2 + 6|\mathcal{R}_{pdf}|^2(21-5\cos2\theta_p)\\ &+\sqrt{6}(3+5\cos2\theta_p)\\ &\times\left[\mathcal{R}_{pdp}\mathcal{R}^*_{pdf} + \mathcal{R}^*_{pdp}\mathcal{R}_{pdf} + 10(\mathcal{R}_{psp}\mathcal{R}^*_{pdf} + \mathcal{R}^*_{psp}\mathcal{R}_{pdf})\right]\\ &+40(\mathcal{R}_{psp}\mathcal{R}^*_{pdp} + \mathcal{R}^*_{psp}\mathcal{R}_{pdp})\Big),\end{aligned}\qquad(6)$$

where $\mathcal{R}_{psp}$, $\mathcal{R}_{pdp}$, and $\mathcal{R}_{pdf}$ correspond to $\mathcal{R}_{l_0 l_\xi l}$ with $l_\xi = 0$ and $l = 1$, $l_\xi = 2$ and $l = 1$, and $l_\xi = 2$ and $l = 3$, respectively.

The process cross section in this case is given by the expression

$$\begin{aligned}\tilde{\sigma}^{(\mathrm{tw})}_{2\gamma,E1}(\theta_c) &= \frac{1}{300}\cos^8\frac{\theta_c}{2}\Big(100|\mathcal{R}_{psp}|^2 + |\mathcal{R}_{pdp}|^2\\ &+36|\mathcal{R}_{pdf}|^2 + 10(\mathcal{R}_{pdp}\mathcal{R}^*_{psp} + \mathcal{R}^*_{pdp}\mathcal{R}_{psp})\Big).\end{aligned}\qquad(7)$$

The cross sections and photoelectron angular distributions corresponding to Eqs. (6) and (7) are shown in Fig. 3.

Discussion

The angular distributions shown in Figs. 2b and 3b look similar. For the single-channel case of helium ionization, the angular distribution is analytically proportional to $\sin^4\theta_p$, however, in neon an additional dependence on the radiation energy appears (see Eq. (6)), and, therefore, on the ratio of the psp, pdp and pdf channel cross sections. But even for a many-electron target such as neon, the wavefunction of an electron emitted by two-photon ionization by a circularly polarized field contains at least $\sin^2\theta_p$, that is why the angular distributions have a similar shape.

The discovered feature of two-photon ionization by Bessel waves of atoms located on the axis of the

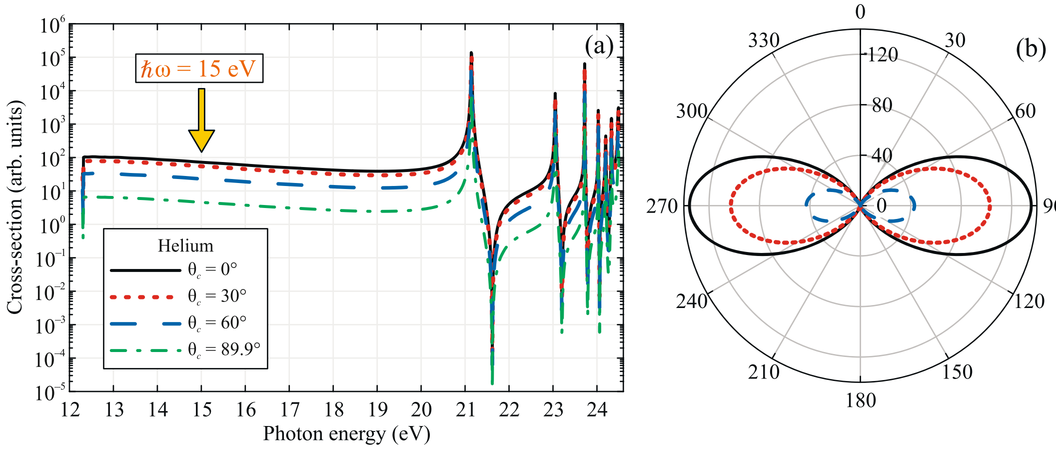

Fig. 2. (Color online) (a) Cross section for two-photon ionization of helium for different opening angles of the Bessel beam cone θ_c. (b) Photoelectron angular distributions corresponding to the incident beam energy $\hbar\omega = 15$ eV for different opening angles of the Bessel beam cone θ_c.

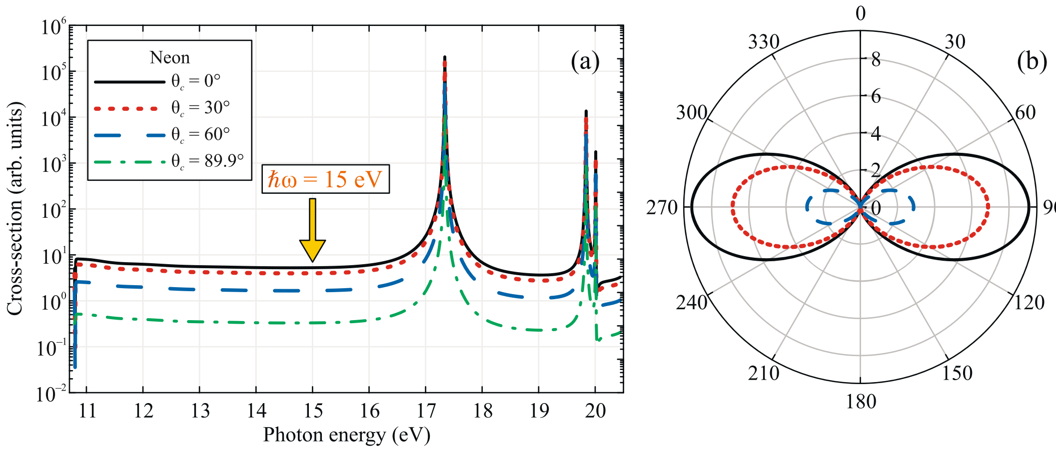

Fig. 3. (Color online) (a) Cross section for two-photon ionization of neon for different opening angles of the Bessel beam cone θ_c. (b) Photoelectron angular distributions corresponding to the incident beam energy $\hbar\omega = 15$ eV for different opening angles of the Bessel beam cone θ_c.

incident beam is the same dependence of both the angular distributions of photoelectrons and the ionization cross sections, regardless of the type of irradiated atoms (or ions), on the opening angle of the Bessel beam cone $\sim\cos^8(\theta_c/2)$, i.e., a part of Eq. (3), summed analytically over L_γ, L'_γ, and k_γ. It is noteworthy that no additional dependencies arise in this case, as a result of which transitions that are "forbidden" in the plane-wave case could become "allowed" (e.g., the $1s \to np \to \epsilon s$ transition in helium). Although such effects could be expected; in particular, it was shown in [28] that the twistedness of the incident radiation leads to the appearance of a component of the photoelectron spin polarization which is forbidden in the process of plane-wave ionization. Thus, the twistedness of the incident radiation in the case of two-photon ionization leads only to a decrease of the process cross section, but not to an increase or the appearance of new qualitative features. It is easy to understand that with an increase in the number N of Bessel waves absorbed by the target atom in the dipole approximation, the dependence of the differential and integral on photoemission angle ionization probabilities on the parameter θ_c will be given by the expression $\sim\cos^{4N}(\theta_c/2)$. Note that although the illustrative calculations were performed within the framework of single active electron approximation and for a pulse of infinite duration, the conclusions presented above are of a general nature. Thus, due to the fact that the geometric part associated with the Bessel field parameter is analytically separated from the energy part, calculations can be performed in an arbitrary model (this applies to the beam structure both in duration and amplitude, as well as to the atomic model used) and all the conclusions made will remain valid.

Analyzing the results presented in Figs. 2 and 3, it becomes clear that for a fixed energy of the incident beam (in the calculations, this energy is taken to be equal to 15 eV for helium and neon), the process cross section can decrease up to 16 times with a maximum increase in the parameter θ_c. The probability of photoemission at a given angle also decreases with increasing θ_c, but the shape of the angular distributions is completely preserved.

3. CONCLUSIONS

The examination of the analytical expression for the angular distribution of the photoelectron emission probability from atoms located on the beam axis and irradiated by Bessel radiation have shown that the angular distributions of two-photon ionization preserve their shape for any parameters of twisted radiation. This statement, proven in the current article, and valid for any multipolarity and polarization of the radiation under consideration, was illustrated by calculations for a circularly polarized field in the dipole approximation. Moreover, the statement is easily generalized to three- and multiphoton transitions, since both the integral probability of the process and the angular dependence are modified by Bessel radiation as $\cos^{4N}(\theta_c/2)$. The proven statement demonstrates the importance of using structured targets, i.e. with a certain distribution of atoms, for observing the effects of radiation twistedness in vector characteristics of multiphoton processes, such as angular distributions.

APPENDIX

DERIVATION OF WORKING EXPRESSIONS

In the formulas below, $\mathcal{J}_n(x)$ is the first kind Bessel function of the order n, $D^k_{lm}(\hat{\mathbf{\Omega}})$ is the Wigner D-function of rotation $\hat{\mathbf{\Omega}}$ on the Euler angle triad, $d^k_{lm}(\theta)$ is the small Wigner D-function, $\hat{a} = \sqrt{2a+1}$, and the standard notation is used for the Clebsch–Gordan coefficients and Wigner $6j$-symbols.

Matrix element of single-photon ionization of an atom by Bessel radiation proceeding according to the scheme

$$\hbar\omega + A(\alpha_i J_i M_i) \to A^+(\alpha_f J_f M_f) + e^-(\mathbf{p} m_s), \quad (A.1)$$

is given by the expression, [29]:

$$\begin{aligned} M^{(\mathrm{tw})}_{\mathrm{cont}} &= \sqrt{\kappa_\perp}\sum_{LMp} i^{L+M-2m_{\mathrm{tam}}}\hat{L}(i\lambda)^p e^{i(m_{\mathrm{tam}}-M)\phi_b} \\ &\times \mathcal{J}_{m_{\mathrm{tam}}-M}(\kappa_\perp b_\perp) d^L_{M\lambda}(\theta_c)\sum_{\kappa\mu J_t M_t}\frac{\hat{l}}{\hat{J}_t}\left(l0,\frac{1}{2}m_s \mid jm_s\right) \quad (A.2) \\ &\times (J_f M_f, j\mu \mid J_t M_t)(J_i M_i, LM \mid J_t M_t) D^{j*}_{\mu m_s}(\hat{\mathbf{p}}) \\ &\times \langle(\alpha_f J_f, \epsilon\kappa)J_t \| H_\gamma(pL) \| \alpha_i J_i\rangle, \end{aligned}$$

where $A(A^+)$ denotes a state before (after) ionization; $\hbar\omega$ is energy of incident photon; $J_{i,f}$ and $M_{i,f}$ are total momentum and its projection for the initial (i) and final (f) states; $\alpha_{i,f}$ is a set of quantum numbers needed to completely describe a state; m_{tam} is a projection of the total angular momentum of a Bessel beam; $\hat{\mathbf{p}} = (\phi_p, \theta_p, 0)$ is direction of photoelectron emission with orbital momentum l and total momentum j; $H_\gamma(pL)$ is the operator of interaction of an atomic electron with a magnetic ($p = 0$) or electric ($p = 1$) photon of multipolarity L. Note that the reduced matrix element $\langle(\alpha_f J_f, \epsilon\kappa)J_t \| H_\gamma(pL) \| \alpha_i J_i\rangle$ includes dependence on the scattering phase (details can also be found in [29]).

Matrix element of single-photon excitation of an atom by Bessel radiation proceeding according to the scheme

$$\hbar\omega + A(\alpha_i J_i M_i) \to A^*(\alpha_f J_f M_f), \quad (A.3)$$

is given by the expression [30]:

$$\begin{aligned} M^{(\mathrm{tw})}_{\mathrm{discrete}} &= \sqrt{\kappa_\perp}\sum_{LMp} i^{L+M-2m_{\mathrm{tam}}}\hat{L}(i\lambda)^p e^{i(m_{\mathrm{tam}}-M)\phi_b} \\ &\times \mathcal{J}_{m_{\mathrm{tam}}-M}(\kappa_\perp b_\perp) d^L_{M\lambda}(\theta_c)\frac{1}{\hat{J}_f}(J_i M_i, LMJ_f|M_f) \quad (A.4) \\ &\times \langle(\alpha_f J_f \| H_\gamma(pL) \| \alpha_i J_i\rangle. \end{aligned}$$

Further, for brevity, we will use $m_{\mathrm{tam}} \equiv m$. In the case of a single atom located on the axis of a Bessel beam ($b_\perp = 0$) and after integration over azimuthal angle ϕ_b ($\int e^{i(m-M)\phi}\frac{d\phi}{2\pi} = \delta_{mM}$) by sequential multiplication of matrix elements (A.2) and (A.4), one can obtain the matrix element of two-photon ionization of an atom by Bessel radiation proceeding according to the scheme shown in Fig. 4, in the form

$$\begin{aligned} M^{(\mathrm{tw})}_{2\gamma} &= \kappa_\perp \sum_{\substack{L_1 L_2 \bar{L}_\gamma \\ J_\xi \mu \bar{M}_\gamma}} i^{L_1+L_2+2m}(i\lambda)^{p_1+p_2}\frac{\hat{L}_1\hat{L}_2\hat{l}\hat{\bar{L}}_\gamma}{\hat{J}_t} \\ &\times (-1)^{-J_0+L_1+L_2-J_t}\left(l0,\frac{1}{2}m_s \mid jm_j\right) D^{j*}_{\mu m_j}(\hat{\mathbf{p}}) \\ &\times (J_0 M_0, \bar{L}_\gamma \bar{M}_\gamma \mid J_t M_t)(J_f M_f, j\mu \mid J_t M_t) \quad (A.5) \\ &\times (L_1 m, L_2 m \mid \bar{L}_\gamma \bar{M}_\gamma) d^{L_1}_{m\lambda}(\theta_c) d^{L_2}_{m\lambda}(\theta_c)\begin{Bmatrix} L_1 & L_2 & \bar{L}_\gamma \\ J_t & J_0 & J_\xi \end{Bmatrix} \\ &\times \langle(\alpha_t J_t \| H_\gamma(p_2 L_2) \| \alpha_\xi J_\xi\rangle\langle(\alpha_\xi J_\xi \| H_\gamma(p_1 L_1) \| \alpha_0 J_0\rangle. \end{aligned}$$

For the convenience of writing further expressions, we introduce the notation

$$\begin{aligned} \langle J_t \| L_1 + L_2 \| J_0\rangle_{2\gamma} &\equiv \sum_{J_\xi} i^{L_1+L_2}(i\lambda)^{p_1+p_2}\begin{Bmatrix} L_1 & L_2 & \bar{L}_\gamma \\ J_t & J_0 & J_\xi \end{Bmatrix} \quad (A.6) \\ &\times \langle(\alpha_t J_t \| H_\gamma(p_2 L_2) \| \alpha_\xi J_\xi\rangle\langle(\alpha_\xi J_\xi \| H_\gamma(p_1 L_1) \| \alpha_0 J_0\rangle. \end{aligned}$$

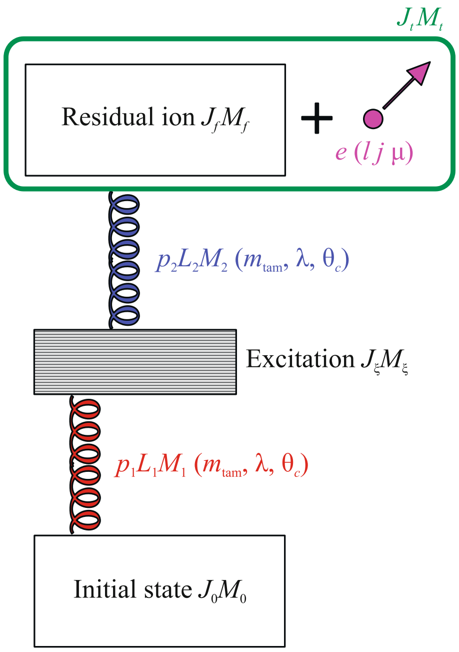

Fig. 4. (Color online) Two-photon ionization scheme. J_0M_0, $J_\xi M_\xi$ and J_fM_f are the total momentum and its projection for the initial state, intermediate bound states (excitations) and the residual ion, respectively. J_tM_t are total momentum and its projection for the system "residual ion + photoelectron."

Note that small Wigner d-functions in Eq. (A.5) can be summed in such a way that the final expression will contain only d-functions with zero projections:

$$\begin{aligned}
d^{L_1}_{m\lambda}(\theta_c)d^{L_2}_{m\lambda}(\theta_c) &= (-1)^{\lambda-m}d^{L_1}_{m\lambda}(\theta_c)d^{L_2}_{-m-\lambda}(\theta_c)\\
&= (-1)^{\lambda-m}\sum_{L_\gamma M_\gamma\Lambda_\gamma}d^{L_\gamma}_{M_\gamma\Lambda_\gamma}(\theta_c)\\
&\times(L_1m,L_2-m\mid L_\gamma M_\gamma)(L_1\lambda,L_2-\lambda\mid L_\gamma\Lambda_\gamma) \qquad \text{(A.7)}\\
&= (-1)^{\lambda-m}\sum_{L_\gamma}d^{L_\gamma}_{00}(\theta_c)\\
&\times(L_1m,L_2-m\mid L_\gamma 0)(L_1\lambda,L_2-\lambda\mid L_\gamma 0).
\end{aligned}$$

The above allows us to rewrite Eq. (A.5) in the form

$$\begin{aligned}
M^{(\rm tw)}_{2\gamma} &= \kappa_\perp\sum_{\substack{L_1L_2L_\gamma\bar{L}_\gamma\\ \bar{M}_\gamma\mu}} i^{2m}\frac{\hat{L}_1\hat{L}_2\hat{l}\hat{\bar{L}}_\gamma}{\hat{J}_t}D^{j^*}_{\mu m_j}(\hat{\mathbf{p}})d^{L_\gamma}_{00}(\theta_c)\\
&\times(-1)^{-J_0+L_1+L_2-J_t+\lambda-m}\left(l0,\frac{1}{2}m_s\mid jm_j\right)\\
&\times(J_0M_0,\bar{L}_\gamma\bar{M}_\gamma\mid J_tM_t)(J_fM_f,j\mu\mid J_tM_t) \qquad \text{(A.8)}\\
&\times(L_1m,L_2-m\mid L_\gamma 0)(L_1\lambda,L_2-\lambda\mid L_\gamma 0)\\
&\times(L_1m,L_2m\mid \bar{L}_\gamma\bar{M}_\gamma)\langle J_t\|L_1+L_2\|J_0\rangle_{2\gamma}.
\end{aligned}$$

The angular distribution of photoelectrons produced in the process of two-photon ionization by Bessel radiation of a single atom located on the axis of the incident beam can be found using basic quantum mechanical principles: $W^{(\rm tw)}_{2\gamma}(\theta_p;\theta_c) = M^{(\rm tw)}_{2\gamma}M^{(\rm tw)*}_{2\gamma}$. Using a matrix element in the form (A.8) and a projection summation technique similar to that used in [29], we obtain a general expression for the angular distribution:

$$\begin{aligned}
&W^{(\rm tw)}_{2\gamma}(\theta_p;\theta_c)\\
&= \sum_{\substack{J_fL_1L_1'L_2L_2'L_\gamma L_\gamma'\\ \bar{L}_\gamma\bar{L}_\gamma'J_tJ_t'll'jj'k_\gamma r}}(-1)^{-J_0-J_f-J_t-J_t'+L_1+L_1'+L_2+L_2'+2j-2m-1/2}\\
&\times\hat{L}_1\hat{L}_1'\hat{L}_2\hat{L}_2'\hat{\bar{L}}_\gamma\hat{\bar{L}}_\gamma'\hat{J}_t\hat{J}_t'\hat{l}\hat{l}'\hat{j}\hat{j}'(L_\gamma0,L_\gamma'0|k_\gamma0)^2\\
&\times(L_1m,L_2m|\bar{L}_\gamma2m)(L_1'm,L_2'm|\bar{L}_\gamma'2m)\\
&\times(L_1m,L_2-m|L_\gamma0)(L_1'm,L_2'-m|L_\gamma'0) \qquad \text{(A.9)}\\
&\times(L_1\lambda,L_2-\lambda|L_\gamma0)(L_1'\lambda,L_2'-\lambda|L_\gamma'0)\\
&\times(\bar{L}_\gamma2m,\bar{L}_\gamma'-2m|r0)(l0,l'0|r0)\\
&\times\begin{Bmatrix} l & l' & r\\ j' & j & \frac{1}{2}\end{Bmatrix}\begin{Bmatrix} j & j' & r\\ J_t' & J_t & J_f\end{Bmatrix}\begin{Bmatrix} \bar{L}_\gamma & \bar{L}_\gamma' & r\\ J_t' & J_t & J_0\end{Bmatrix}\\
&\times P_r(\cos\theta_p)P_{k_\gamma}(\cos\theta_c)\\
&\times\langle J_t\|L_1+L_2\|J_0\rangle_{2\gamma}\langle J_t'\|L_1'+L_2'\|J_0\rangle^*_{2\gamma}.
\end{aligned}$$

As an illustration, this paper considers ionization of $1s$ shell of helium atom and $2p$ shell of neon atom in neutral states ($J_0=0$) in the electric dipole approximation ($L_{1,2}=L_{1,2}'=1$; $p_{1,2}=1$). In this case $(j\pm1/2)$ and J_ξ are integers, therefore, Eq. (A.9) is simplified and takes the form

$$\begin{aligned}
W^{(\rm tw)}_{2\gamma,E1}(\theta_p;\theta_c) &= 9\times\sum_{\substack{J_fJ_tJ_t'L_\gamma L_\gamma'\\ \bar{L}_\gamma\bar{L}_\gamma'll'jj'k_\gamma r}}(-1)^{-J_f-J_t-J_t'+1/2}\\
&\times\hat{\bar{L}}_\gamma\hat{\bar{L}}_\gamma'\hat{J}_t\hat{J}_t'\hat{l}\hat{l}'\hat{j}\hat{j}'(L_\gamma0,L_\gamma'0\mid k_\gamma0)^2(11,1-1\mid L_\gamma0)^2\\
&\times(11,1-1\mid L_\gamma'0)^2(11,11\mid\bar{L}_\gamma2)(11,11\mid\bar{L}_\gamma'2) \qquad \text{(A.10)}\\
&\times(\bar{L}_\gamma2,\bar{L}_\gamma'-2\mid r0)(l0,l'0\mid r0)\\
&\times\begin{Bmatrix} l & l' & r\\ j' & j & \frac{1}{2}\end{Bmatrix}\begin{Bmatrix} j & j' & r\\ J_t' & J_t & J_f\end{Bmatrix}\begin{Bmatrix} \bar{L}_\gamma & \bar{L}_\gamma' & r\\ J_t' & J_t & 0\end{Bmatrix}\\
&\times P_r(\cos\theta_p)P_{k_\gamma}(\cos\theta_c)\mathcal{A}^{E1}_{2\gamma}\mathcal{A}^{E1*}_{2\gamma},
\end{aligned}$$

where

$$\begin{aligned}
\mathcal{A}^{E1}_{2\gamma} &= \sum_{J_\xi}(-1)^{J_\xi}\begin{Bmatrix} J1 & 1 & \bar{L}_\gamma\\ J_t & 0 & J_\xi\end{Bmatrix} \qquad \text{(A.11)}\\
&\times\langle(\alpha_tJ_t\|H_\gamma(E1)\|\alpha_\xi J_\xi\rangle\langle(\alpha_\xi J_\xi\|H_\gamma(E1)\|\alpha_0J_0\rangle.
\end{aligned}$$

Next, we will use the single-active-electron model, representing the initial system as a set of the ion core and the ionized (active) electron. To do this, in (A.11) we will separate the moments according to the scheme:

$$\begin{aligned} J_0 &= L_0(L_f l_0) + S_0 \\ J_\xi &= L_\xi(L_f l_\xi) + S_\xi \\ J_t &= L_t(L_f l) + S_t, \end{aligned} \tag{A.12}$$

where L_f is an orbital angular momentum of the ion core (residual ion); l_0, l_ξ and l are orbital momenta of an ionized electron in the initial state of an atom, in an excited state, and in a continuum, respectively.

Taking into account that $J_f = L_f + S_f$, $S_0 = S_\xi = S_t = 0$, $S_f = 1/2$, $J_\xi = L_\xi = 1$ and, as a consequence, $\bar{L}_\gamma = J_t = L_t$, and also using Eqs. (A.72) and (A.73) from [31], it is easy to show that the amplitude (A.11) is reduced to the form

$$\begin{aligned} \widetilde{\mathcal{A}}^{E1}_{2\gamma} &= \frac{1}{\sqrt{2}} \sum_{l_\xi} (-1)^{j+l_0+\bar{L}_\gamma-1/2} \frac{\hat{J}_f \hat{j}}{\hat{L}_f} \begin{Bmatrix} J_f & L_f & \frac{1}{2} \\ l & j & \bar{L}_\gamma \end{Bmatrix} \\ &\times \begin{Bmatrix} L_f & l & \bar{L}_\gamma \\ 1 & 1 & l_\xi \end{Bmatrix} \langle l \| H_\gamma(E1) \| l_\xi \rangle \langle l_\xi \| H_\gamma(E1) \| l_0 \rangle. \end{aligned} \tag{A.13}$$

Then, in the one-electron model, the expression for the angular distribution (A.10) takes the form

$$\begin{aligned} \widetilde{W}^{(\mathrm{tw})}_{2\gamma,E1}(\theta_p;\theta_c) &= \frac{9}{2} \times \sum_{\substack{L_\gamma L'_\gamma \bar{L}_\gamma \bar{L}'_\gamma \\ J_f l_\xi l'_\xi l l' j j' k_\gamma r}} (-1)^{j+j'-J_f-1/2} \\ &\times \frac{\hat{\bar{L}}^2_\gamma \hat{\bar{L}}'^2_\gamma \hat{J}_f^{\,2} \hat{j}^2 \hat{j}'^2 \hat{l} \hat{l}'}{\hat{L}_f^2} (L_\gamma 0, L'_\gamma 0 | k_\gamma 0)^2 (11, 1-1 | L_\gamma 0)^2 \\ &\times (11, 1-1 \mid L'_\gamma 0)^2 (11, 11 \mid \bar{L}_\gamma 2)(11, 11 \mid \bar{L}'_\gamma 2) \\ &\times (\bar{L}_\gamma 2, \bar{L}_\gamma - 2 \mid r0)(l0, l'0 \mid r0) \\ &\times \begin{Bmatrix} J_f & L_f & \frac{1}{2} \\ l' & j' & \bar{L}'_\gamma \end{Bmatrix} \begin{Bmatrix} L_f & l & \bar{L}_\gamma \\ 1 & 1 & l_\xi \end{Bmatrix} \\ &\times \begin{Bmatrix} L_f & l & \bar{L}_\gamma \\ 1 & 1 & l_\xi \end{Bmatrix} \begin{Bmatrix} L_f & l' & \bar{L}'_\gamma \\ 1 & 1 & l'_\xi \end{Bmatrix} \\ &\times \begin{Bmatrix} j & j' & r \\ \bar{L}'_\gamma & \bar{L}_\gamma & J_f \end{Bmatrix} \begin{Bmatrix} \bar{L}_\gamma & \bar{L}_\gamma & r \\ \bar{L}'_\gamma & \bar{L}_\gamma & 0 \end{Bmatrix} \\ &\times P_r(\cos\theta_p) P_{k_\gamma}(\cos\theta_c) \mathcal{R}_{l_0 l_\xi l} \mathcal{R}^*_{l_0 l'_\xi l'}, \end{aligned} \tag{A.14}$$

where $\mathcal{R}_{l_0 l_\xi l} \equiv \langle l \| H_\gamma(E1) \| l_\xi \rangle \langle l_\xi \| H_\gamma(E1) \| l_0 \rangle$.

After taking the sum on j, j' and J_f we finally get:

$$\begin{aligned} \widetilde{W}^{(\mathrm{tw})}_{2\gamma,E1}(\theta_p;\theta_c) &= 9 \times \sum_{\substack{L_\gamma L'_\gamma \bar{L}_\gamma \bar{L}'_\gamma \\ l_\xi l'_\xi l l' k_\gamma r}} (-1)^{L_f+l+l'} \frac{\hat{\bar{L}}_\gamma \hat{\bar{L}}'_\gamma \hat{l} \hat{l}'}{\hat{L}_f^{\,2}} \\ &\times (11, 1-1 | L_\gamma 0)^2 (11, 1-1 | L'_\gamma 0)^2 (L_\gamma 0, L'_\gamma 0 | k_\gamma 0)^2 \\ &\times (11, 11 | \bar{L}_\gamma 2)(11, 11 | \bar{L}'_\gamma 2)(\bar{L}_\gamma 2, \bar{L}_\gamma - 2 | r0)(l0, l'0 | r0) \\ &\times \begin{Bmatrix} L_f & l & \bar{L}_\gamma \\ 1 & 1 & l_\xi \end{Bmatrix} \begin{Bmatrix} L_f & l' & \bar{L}'_\gamma \\ 1 & 1 & l'_\xi \end{Bmatrix} \begin{Bmatrix} l & l' & r \\ \bar{L}'_\gamma & \bar{L}_\gamma & L_f \end{Bmatrix} \\ &\times P_r(\cos\theta_p) P_{k_\gamma}(\cos\theta_c) \mathcal{R}_{l_0 l_\xi l} \mathcal{R}^*_{l_0 l'_\xi l'}. \end{aligned} \tag{A.15}$$

If we consider the circularly polarized incident field, then, taking into account the dipole approximation in (A.15), only $\bar{L}_\gamma = \bar{L}'_\gamma = 2$ remain, i.e.:

$$\begin{aligned} \widetilde{W}^{(\mathrm{tw,circ})}_{2\gamma,E1}(\theta_p;\theta_c) &= 45 \times \sum_{\substack{L_\gamma L'_\gamma k_\gamma r \\ l_\xi l'_\xi l l'}} (-1)^{L_f+l+l'} \frac{\hat{l} \hat{l}'}{\hat{L}_f^2} \\ &\times (11, 1-1 | L_\gamma 0)^2 (11, 1-1 | L'_\gamma 0)^2 (L_\gamma 0, L'_\gamma 0 | k_\gamma 0)^2 \\ &\times (22, 2-2 \mid r0)(l0, l'0 \mid r0) \\ &\times \begin{Bmatrix} L_f & l & 2 \\ 1 & 1 & l_\xi \end{Bmatrix} \begin{Bmatrix} L_f & l' & 2 \\ 1 & 1 & l'_\xi \end{Bmatrix} \\ &\times \begin{Bmatrix} l & l' & r \\ 2 & 2 & L_f \end{Bmatrix} \times P_r(\cos\theta_p) P_{k_\gamma}(\cos\theta_c) \mathcal{R}_{l_0 l_\xi l} \mathcal{R}^*_{l_0 l'_\xi l'}. \end{aligned} \tag{A.16}$$

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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