Absorption of twisted light by hydrogenlike atoms

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Theoretical analysis of the excitation of low-Z hydrogenlike atoms by incident twisted light is presented. Emphasis is placed on the cross sections that describe transitions between particular atomic substates. Simple expressions for these *partial* cross sections are derived using the nonrelativistic first-order perturbation theory and the momentum representation of the photon wave functions. Based on the developed approach, calculations have been performed for the $1s \to 2p$ and $2p \to 3d$ transitions induced in the course of interaction of twisted (Bessel) light with a macroscopic hydrogen target. Results of these calculations, supported by an analytical analysis, clearly indicate that the sublevel population of residual atoms following absorption of twisted photons differs much from what is expected for the standard plane-wave case; this effect can be easily observed experimentally by measuring the linear polarization of the subsequent fluorescent emission.

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I. INTRODUCTION

With the recent experimental advances in optics and photonics, new possibilities arise to design light beams with special polarization and angular momentum properties, unusual for the "standard" plane-wave radiation. Of particular interest here are *twisted* photons that carry a nonzero projection of the orbital angular momentum (OAM) onto their propagation direction [1,2]. Such photons are readily produced with the help of computer-generated holograms, spiral phase plates, and helical undulators [3], and can serve as a novel tool to explore the role of the OAM in fundamental light-matter interaction processes. During the last few years, for example, a number of theoretical proposals have been made to employ the twisted light in photoionization as well as Compton and elastic (photon-atom) scattering studies [4–7]. These investigations demonstrate that the properties of emitted and/or scattered particles may be strongly affected by the OAM and the phase structure of the incident radiation.

Besides the ionization and scattering processes, much of the recent interest has been focused also on the *excitation* of target atoms by twisted photons. Detailed theoretical analysis of the bound-state transitions in hydrogenlike ions was performed by Afanasev and co-workers [8–10] who used the coordinate representation of the photon wave functions to evaluate the transition matrix elements and to obtain the general selection rules. In the present work we propose an alternative approach to the treatment of the atomic photoexcitation process. By performing a nonrelativistic study in the *momentum* space we develop a simpler formalism for the evaluation of the transition cross sections as well as properties of the excited atomic states and subsequent fluorescent radiation. Our results, moreover, can be employed for the guidance and analysis of future experimental investigations of the interaction of the twisted photons beams with atomic ensembles.

To lay down a general theory for the description of the excitation of hydrogenlike atoms by twisted light we start in Sec. II A with the evaluation of the bound-state transition amplitudes. We show, in particular, how the matrix elements for the absorption of twisted (Bessel) photons can be expressed in terms of their "plane-wave" analogs. In Sec. II B we employ these matrix elements to derive the *partial* cross sections that describe photoexcitation into particular atomic substates $|n\,l\,m\rangle$. Two scenarios are considered here, in which the Bessel beam collides with either a well-localized single atom or a macroscopic (atomic) target. Detailed calculations were performed for the second, more experimentally realistic case and are presented in Sec. III. Based on the results obtained for the $1s \to 2p$ and $2p \to 3d$ transitions, we show that the absorption of Bessel light may lead to the population of atomic substates which otherwise cannot be reached by the standard plane-wave excitation. It is shown, moreover, that the kinematic properties of the incident twisted photons, such as the ratio of the transverse to longitudinal momenta, can also influence the linear polarization of the subsequent fluorescent radiation which can be easily detected experimentally. A brief summary of these results and an outlook are given finally in Sec. IV. Hartree atomic units ($\hbar = e = m_e = 1$, $c = 1/\alpha$) are used throughout the paper unless stated otherwise.

II. THEORY

A. Evaluation of transition amplitude

1. Plane-wave photons

Within the framework of the (nonrelativistic) perturbation theory, the photoexcitation of a hydrogenlike atom from an initial $|i\rangle = |n_i l_i m_i\rangle$ to a final $|f\rangle = |n_f l_f m_f\rangle$ bound state is described by the matrix element:

$$M_{m_f m_i} = \int \psi^*_{n_f l_f m_f}(\boldsymbol{r})\hat{V}\psi_{n_i l_i m_i}(\boldsymbol{r})\,d\boldsymbol{r}. \qquad (1)$$

Here we assume that the nuclear charge Z of a target atom is relatively low and, hence, the standard Schrödinger wave

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functions $\psi_{n_i l_i m_i}(\boldsymbol{r}) = R_{n_i l_i}(r)\, Y_{l_i m_i}(\theta,\phi)$ and $\psi_{n_f l_f m_f}(\boldsymbol{r}) = R_{n_f l_f}(r)\, Y_{l_f m_f}(\theta,\phi)$ can be employed. In Eq. (1), moreover, $\hat{V}$ is the transition operator which describes the interaction of an electron with the electromagnetic field. The explicit form of this operator depends on the state in which the incident light is *prepared*. For a plane-wave photon with the wave vector $\boldsymbol{k}$, energy $\omega = k/\alpha$, and helicity $\lambda = \pm 1$, for example, the (nonrelativistic) transition operator reads

$$\hat{V}^{(\mathrm{pl})} = \alpha\ \mathbf{A}_{\boldsymbol{k}\lambda}(\boldsymbol{r})\, \hat{\boldsymbol{p}} = \alpha\, \boldsymbol{e}_{\boldsymbol{k}\lambda}\, e^{i\boldsymbol{k}\boldsymbol{r}}\, \hat{\boldsymbol{p}}, \tag{2}$$

where $\hat{\boldsymbol{p}} = -i\hat{\boldsymbol{\nabla}}$ is the linear momentum operator and $\boldsymbol{e}_{\boldsymbol{k}\lambda}$ denotes the polarization vector.

Computation of the matrix element (1) with the plane-wave operator (2) also requires us to specify the quantization (z) axis of the overall system. In atomic studies, the quantization axis is often adopted along the wave vector of the incoming radiation, $z \,||\, \boldsymbol{k}$. This choice of the coordinate system, together with the fact that the photon polarization and wave vector are orthogonal to each other, $\boldsymbol{k}\cdot\boldsymbol{e}_{\boldsymbol{k}\lambda} = 0$, allows one to simplify the matrix element (1) into

$$\begin{aligned} &M^{(\mathrm{pl})}_{m_f m_i}(\theta_k = 0, \phi_k = 0) \\ &\quad \equiv M^{(\mathrm{pl})}_{m_f m_i}(0,0) \\ &\quad = -i\alpha \int \psi^*_{n_f l_f m_f}(\boldsymbol{r})\, e^{ikz} \nabla_\lambda\, \psi_{n_i l_i m_i}(\boldsymbol{r})\, d\boldsymbol{r}, \end{aligned} \tag{3}$$

where ∇_λ refers to the spherical components of the nabla operator, and (θ_k,ϕ_k) are the polar and azimuthal vectors of the wave vector $\boldsymbol{k}$.

To further evaluate the amplitude (3) we use the Rayleigh expansion of the photon plane wave in terms of spherical Bessel functions $j_L(kr)$:

$$e^{i\,k\,z} = \sqrt{4\pi} \sum_L i^L \sqrt{2L+1}\, j_L(kr)\, Y_{L0}(\theta,\phi), \tag{4}$$

and the formula for the gradient of the initial wave function [11]:

$$\begin{aligned} \nabla_\lambda\, \psi_{n_i l_i m_i}(\boldsymbol{r}) &\equiv \nabla_\lambda \big(R_{n_i l_i}(r)\, Y_{l_i m_i}(\theta,\phi) \big) \\ &= \sum_{\Lambda_i = l_i \pm 1} A^\lambda_{\Lambda_i m_i}\, \tilde{R}_{n_i \Lambda_i}(r)\, Y_{\Lambda_i\, m_i+\lambda}(\theta,\phi). \end{aligned} \tag{5}$$

In the latter expression, the coefficients $A^\lambda_{\Lambda_i m_i}$ read as

$$\begin{aligned} A^{\pm 1}_{\Lambda_i m_i} &= \sqrt{\frac{(l_i \pm m_i + 1)(l_i \pm m_i + 2)}{2(2l_i+1)(2l_i+3)}}, \quad \text{if } \Lambda_i = l_i + 1, \\ A^{\pm 1}_{\Lambda_i m_i} &= -\sqrt{\frac{(l_i \mp m_i - 1)(l_i \mp m_i)}{2(2l_i-1)(2l_i+1)}}, \quad \text{if } \Lambda_i = l_i - 1, \end{aligned} \tag{6}$$

and the radial functions $\tilde{R}_{n_i\Lambda_i}(r)$ are given by

$$\begin{aligned} \tilde{R}_{n_i\Lambda_i} &= \left(\frac{\partial R_{n_i l_i}(r)}{\partial r} - \frac{l_i}{r} R_{n_i l_i}(r) \right), \quad \text{if } \Lambda_i = l_i + 1, \\ \tilde{R}_{n_i\Lambda_i} &= \left(\frac{\partial R_{n_i l_i}(r)}{\partial r} + \frac{l_i+1}{r} R_{n_i l_i}(r) \right), \quad \text{if } \Lambda_i = l_i - 1. \end{aligned} \tag{7}$$

By inserting decompositions (4) and (5) into Eq. (3) and making some angular momentum algebra, we obtain the final expression for the matrix element:

$$\begin{aligned} M^{(\mathrm{pl})}_{m_f m_i}(0,0) = -i\alpha \sum_{L\Lambda_i} i^L \sqrt{\frac{(2\Lambda_i+1)(2L+1)^3}{(2l_f+1)}} A^\lambda_{\Lambda_i m_i} \\ \times \langle \Lambda_i 0\ L0 | l_f 0 \rangle \langle \Lambda_i m_i + \lambda\, L0 | l_f m_f \rangle \\ \times \int_0^\infty R_{n_f l_f}(r)\, j_L(kr)\, \tilde{R}_{n_i\Lambda_i}(r)\, r^2\, dr, \end{aligned} \tag{8}$$

which describes the excitation of a hydrogenlike system if a plane-wave photon is absorbed. The symmetry properties of the Clebsch-Gordan coefficients $\langle \ldots | \ldots \rangle$ in this equation immediately lead to the well-known selection rule:

$$m_i + \lambda = m_f, \quad l_f \geqslant |m_f|, \tag{9}$$

which defines the sublevel population of the residual atom. In the present work, the matrix element (8) was calculated by means of the DIRAC program [12,13], a computer-algebraic toolbox which was developed by us for studying the properties and the dynamical behavior of hydrogenlike ions.

The transition amplitude (8) has been derived under the assumption that the incident light propagates along the (z) quantization axis. Such an assumption is usually well justified to explore the interaction of atoms with the *plane-wave* radiation, where the photon wave vector $\boldsymbol{k}$ is the only preferred direction of the overall system. In the next section, however, we will see that for *twisted* photons one needs the matrix element (1) with the operator (2) in which the wave vector $\boldsymbol{k}$ points in an arbitrary direction, $\hat{\boldsymbol{k}} = \boldsymbol{k}/k = (\theta_k,\phi_k)$, not coinciding with the quantization z- axis. To calculate the transition amplitude $M^{(\mathrm{pl})}_{fi}(\theta_k,\phi_k)$ for this general case, we express the (initial- and final-state) atomic wave functions, defined in the coordinate system $S(x,y,z)$, in terms of the functions from the system $S(x',y',z')$ with the z' axis along the vector $\boldsymbol{k}$. Since the $S(x',y',z')$ coordinate system is obtained from the $S(x,y,z)$ by a *rotation* through an angle θ_k around the y axis and an angle ϕ_k around the z axis, one obtains

$$\psi_{nlm}(\boldsymbol{r}) = \sum_{m'} D^{l*}_{mm'}(\phi_k,\theta_k,0) \psi_{nlm'}(\boldsymbol{r}'), \tag{10}$$

where $D^l_{mm'}(\phi_k,\theta_k,0) = e^{-im\phi_k}\, d^l_{mm'}(\theta_k)$ is the Wigner D function, and $d^l_{mm'}(\theta_k)$ can be written in terms of polynomials in the sine and cosine of $\theta_k/2$, see Ref. [11]. By inserting this expression into Eq. (1), we can derive

$$\begin{aligned} &M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,\phi_k) \\ &\quad = -i\alpha \int \psi^*_{n_f l_f m_f}(\boldsymbol{r})\, \boldsymbol{e}_{\boldsymbol{k}\lambda}\, e^{i\boldsymbol{k}\boldsymbol{r}}\, \hat{\boldsymbol{\nabla}}\, \psi_{n_i l_i m_i}(\boldsymbol{r})\, d\boldsymbol{r} \\ &\quad = -i\alpha \sum_{m'_f m'_i} D^{l_f}_{m_f m'_f}(\phi_k,\theta_k,0)\, D^{l_i *}_{m_i m'_i}(\phi_k,\theta_k,0) \\ &\qquad \times \int \psi^*_{n_f l_f m'_f}(\boldsymbol{r}')\, e^{ikz'}\, \nabla_\lambda\, \psi_{n_i l_i m'_i}(\boldsymbol{r}')\, d\boldsymbol{r}' \\ &\quad = e^{-i(m_f - m_i)\phi_k} \sum_{m'_f m'_i} d^{l_f}_{m_f m'_f}(\theta_k)\, d^{l_i}_{m_i m'_i}(\theta_k) M^{(\mathrm{pl})}_{m'_f m'_i}(0,0). \end{aligned} \tag{11}$$

Here m_i' and m_f' are projections of the initial- and final-state angular momenta on the propagation direction of incident light, i.e., on the z' axis.

Equations (8)–(11) can be further simplified for the excitation of a hydrogenlike atom from its ground $1s$ state, i.e., when $n_i = 1$ and $l_i = m_i = 0$. In this case, the $M^{(\mathrm{pl})}_{m_f m_i}(0,0)$ reads as

$$M^{(\mathrm{pl})}_{m_f,\,m_i=0}(0,0) = -i\alpha \sum_L i^L \sqrt{\frac{(2L+1)^3}{2l_f+1}} \langle 10\, L0|l_f 0\rangle\, \langle 1\lambda\, L0|l_f m_f\rangle \times \int_0^\infty R_{n_f l_f}(r) j_L(kr) \frac{\partial R_{10}(r)}{\partial r}\, r^2\, dr, \tag{12}$$

where, owing to the properties of the Clebsch-Gordan coefficients, the summation is restricted to $L = l_f \pm 1$ and $m_f = \lambda$. By inserting this expression into Eq. (11) we find the matrix element for the transition $|1s\rangle + \gamma \to |n_f\, l_f\, m_f\rangle$ and for an arbitrary direction of light propagation:

$$M^{(\mathrm{pl})}_{m_f,\,m_i=0}(\theta_k,\phi_k) = D^{l_f}_{m_f m_f'}(\phi_k,\theta_k,0) M^{(\mathrm{pl})}_{\lambda,\,m_i'=0}(0,0) = e^{-im_f\phi_k}\, d^{l_f}_{m_f\lambda}(\theta_k)\, M^{(\mathrm{pl})}_{\lambda,\,m_i'=0}(0,0). \tag{13}$$

As seen from this formula, the $M^{(\mathrm{pl})}_{m_f,\,m_i=0}(\theta_k,\phi_k)$ is trivially factorized into the product of the Wigner D matrix $D^{l_f}_{m_f m_f'}(\phi_k,\theta_k,0)$, which depends on the incident photon angles (θ_k,ϕ_k), and the plane-wave amplitude (12). Below, this factorization will allow us to perform an *analytical* analysis of the absorption of the twisted light.

2. Twisted photons

To derive the amplitude for the excitation of hydrogenlike atoms by twisted radiation we have to come back to the general expression (1) and to take the transition operator as

$$\hat{V}^{(\mathrm{tw})} = \alpha\, \mathbf{A}_{\varkappa m_\gamma k_z \lambda}(\boldsymbol{r})\, \hat{\boldsymbol{p}}. \tag{14}$$

This operator is similar to the plane-wave one (2), except for the vector potential $\mathbf{A}_{\varkappa m_\gamma k_z\lambda}$ which describes here the (twisted) wave propagating along the quantization axis with well-defined longitudinal momentum k_z and the projection of the total angular momentum (TAM), $J_z = m_\gamma$. We further assume that the absolute value of the transverse momentum, $|\boldsymbol{k}_\perp| = \varkappa$, and, hence, the energy of the photons, $\omega = k/\alpha = \sqrt{k_z^2 + \varkappa^2}/\alpha$, are also fixed. Such a *Bessel* state of light is characterized by the vector potential [6,14]

$$\mathbf{A}_{\varkappa m_\gamma k_z\lambda}(\boldsymbol{r}) = \int \boldsymbol{e}_{\boldsymbol{k}\lambda}\, e^{i\boldsymbol{k}\boldsymbol{r}}\, a_{\varkappa m_\gamma}(\boldsymbol{k}_\perp)\, e^{-i\boldsymbol{k}_\perp\boldsymbol{b}}\, \frac{d^2k_\perp}{(2\pi)^2}, \tag{15}$$

which can be written as a superposition (integral) of the standard plane-wave components with the amplitude

$$a_{\varkappa m_\gamma}(\boldsymbol{k}_\perp) = (-i)^{m_\gamma}\, e^{im_\gamma\phi_k} \sqrt{\frac{2\pi}{k_\perp}}\, \delta(k_\perp - \varkappa). \tag{16}$$

In Eq. (15), moreover, the factor $e^{-i\boldsymbol{k}_\perp\boldsymbol{b}}$ specifies the position of a target atom within the incident wave front, where $\boldsymbol{b}$ is the impact parameter vector shown in Fig. 1. This exponential factor is essential since in contrast to a plane wave the twisted beam has a complex spatial structure. Within a plane perpendicular to the propagation (z) axis, in particular, the intensity profile of the Bessel radiation exhibits the concentric ring pattern with a central zero-intensity spot [6]. In the analysis below we will define the impact parameter $\boldsymbol{b} = (b_x,b_y,0)$ with regard to such a (zero-intensity) center.

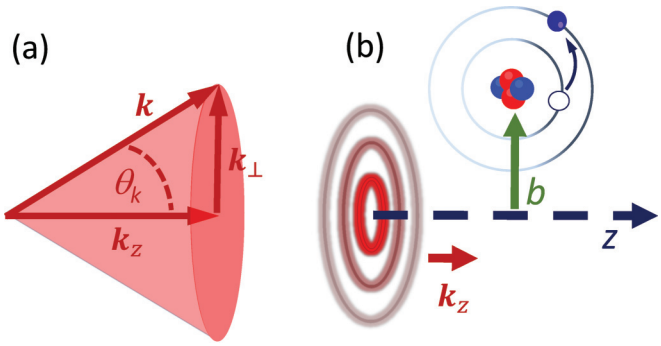

FIG. 1. (Color online) Left panel: In the momentum space, the twisted light can be seen as a coherent superposition of plane waves with wave vectors $\boldsymbol{k}$, lying on a cone with a polar opening angle $\theta_k = \arctan(|\boldsymbol{k}_\perp|/k_z)$, and with polarization vectors $\boldsymbol{e}_{\boldsymbol{k}\lambda}$, perpendicular to $\boldsymbol{k}$. Right panel: Geometry of the atomic excitation by twisted Bessel light. The quantization (z) axis is chosen along the propagation direction of the incident beam. The position of a target atom with respect to the zero-intensity line of the Bessel wave is characterized by the impact parameter b.

By inserting the interaction operator (14)–(16) into Eq. (1) and performing trivial integration over the transverse momentum $k_\perp$, we can write the amplitude for the excitation of an atom by twisted light as

$$M^{(\mathrm{tw})}_{m_f m_i}(\boldsymbol{b}) = \int M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,\varphi_k)\, a_{\varkappa m_\gamma}(\boldsymbol{k}_\perp)\, e^{-i\boldsymbol{k}_\perp\boldsymbol{b}}\, \frac{d^2k_\perp}{(2\pi)^2} = (-i)^{m_\gamma} \sqrt{\frac{\varkappa}{2\pi}} \int_0^{2\pi} \frac{d\varphi_k}{2\pi}\, e^{im_\gamma\varphi_k - i\boldsymbol{k}_\perp\boldsymbol{b}} \times M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,\varphi_k), \tag{17}$$

where $\boldsymbol{k} = (\varkappa\cos\varphi_k, \varkappa\sin\varphi_k, k_z)$ and $\tan\theta_k = \varkappa/k_z$. If we use the well-known integral relation

$$\int_0^{2\pi} \frac{d\varphi}{2\pi}\, e^{in\varphi \pm iz\cos\varphi} = (\pm i)^n\, J_n(z), \tag{18}$$

together with Eq. (11) and the scalar product formula $\boldsymbol{k}_\perp\boldsymbol{b} = \varkappa\, b\, \cos(\varphi_k - \varphi_b)$, we finally obtain the expression

$$M^{(\mathrm{tw})}_{m_f m_i}(\boldsymbol{b}) = i^{m_f - m_i - 2m_\gamma}\, e^{i(m_\gamma + m_i - m_f)\varphi_b} \sqrt{\frac{\varkappa}{2\pi}} \times J_{m_\gamma + m_i - m_f}(\varkappa b) \sum_{m_f' m_i'} d^{l_f}_{m_f m_f'}(\theta_k)\, d^{l_i}_{m_i m_i'}(\theta_k) \times M^{(\mathrm{pl})}_{m_f' m_i'}(0,0). \tag{19}$$

This formula shows that the amplitude $M^{(\mathrm{tw})}_{m_f m_i}(\boldsymbol{b})$ for the twisted light can be expressed in terms of the matrix elements (3) for the absorption of plane-wave photons, propagating along the quantization axis. For the excitation from

the ground $1s$ state, especially, this expression shows that $M^{(\mathrm{tw})}_{m_f m_i}(\boldsymbol{b})$ and $M^{(\mathrm{pl})}_{m_f m_i}(0,0)$ are simply *proportional* to each other:

$$M^{(\mathrm{tw})}_{m_f\, m_i=0}(\boldsymbol{b}) = i^{\,m_f-2m_\gamma}\, e^{i(m_\gamma-m_f)\varphi_b}\sqrt{\frac{\varkappa}{2\pi}}\,J_{m_\gamma-m_f}(\varkappa b) \times d^{\,l_f}_{m_f\lambda}(\theta_k)\, M^{(\mathrm{pl})}_{\lambda\, m_i'=0}(0,0), \tag{20}$$

and the proportionality coefficient only depends on the parameters of the incident twisted radiation.

B. Photoexcitation cross sections

We can apply the matrix elements (3), (11), and (19) to calculate the cross section for the excitation of an atom into a particular sublevel $|n_f\, l_f\, m_f\rangle$. For the incident plane-wave radiation with flux $j = k/(2\pi)$, this *partial* excitation cross section is simply given by:

$$\sigma^{(\mathrm{pl})}_{m_f} = \frac{1}{2l_i+1}\frac{2\pi}{j}\sum_{m_i}\left|M^{(\mathrm{pl})}_{m_f m_i}(0,0)\right|^2\delta(\omega+E_i-E_f), \tag{21}$$

if the atom is assumed to be initially unpolarized. Moreover, since we restrict our analysis to photons with certain helicity λ, the summation in Eq. (21) is restricted to $m_i = m_f - \lambda$.

By using the amplitude $M^{(\mathrm{tw})}_{m_f m_i}$ one can derive the excitation cross section for the incoming twisted (Bessel) beam of radius R. In contrast to the plane-wave case, we need first to agree about the particular setup under which the photoabsorption process is observed. For a well-defined impact parameter of an atom with regard to the photon beam axis, for example, the partial cross section reads

$$\begin{aligned}\sigma^{(\mathrm{tw})}_{m_f}(\boldsymbol{b}) &= \frac{2\pi}{j_z}\frac{1}{2l_i+1}\sum_{m_i}\left|M^{(\mathrm{tw})}_{m_f m_i}(\boldsymbol{b})\right|^2\delta(\omega+E_i-E_f)\\ &= \frac{2\pi}{j_z}\frac{\varkappa}{2\pi}\frac{1}{2l_i+1}\,\delta(\omega+E_i-E_f)\\ &\quad\times\sum_{m_i}\left|J_{m_\gamma+m_i-m_f}(\varkappa b)\, M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,0)\right|^2,\end{aligned} \tag{22}$$

where we employed Eqs. (11) and (19), and introduced the averaged flux $j_z = k_z/(2\pi^3 R)$ of the Bessel radiation (see for details Appendix B in Ref. [14]). In practice, however, no accurate knowledge about the position of an individual atom within the wave front is usually available. As a second scenario, therefore, we consider the collision of the Bessel photons with a target in which atoms are distributed randomly over the extent of the incident beam. For such a *macroscopic* target we have to average the excitation cross section over the impact parameters $b \leqslant R$. By employing the first line of Eq. (17) and definition (16), the integration over the $\boldsymbol{b}$ and the transverse momentum $\boldsymbol{k}_\perp$ is trivially performed to give

$$\begin{aligned}\sigma^{(\mathrm{tw})}_{m_f}(\theta_k) &= \frac{2\pi}{j_z}\,\delta(\omega+E_i-E_f)\frac{1}{2l_i+1}\sum_{m_i}\int\left|M^{(\mathrm{tw})}_{m_f m_i}(\boldsymbol{b})\right|^2\frac{d^2b}{\pi R^2} = \frac{2\pi}{j_z}\,\delta(\omega+E_i-E_f)\frac{1}{2l_i+1}\\ &\quad\times\sum_{m_i}\int e^{i(\boldsymbol{k}'_\perp-\boldsymbol{k}_\perp)\boldsymbol{b}}\, a_{\varkappa m_\gamma}(\boldsymbol{k}_\perp)\, a^*_{\varkappa m_\gamma}(\boldsymbol{k}'_\perp)\, M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,\varphi_k) M^{\mathrm{pl}*}_{m'_f m_i}(\theta'_k,\varphi'_k)\frac{d^2k_\perp}{(2\pi)^2}\frac{d^2k'_\perp}{(2\pi)^2}\frac{d^2b}{\pi R^2}\\ &= \frac{2}{j_z R^2}\,\delta(\omega+E_i-E_f)\frac{1}{2l_i+1}\sum_{m_i}\int\left|\delta(k_\perp-\varkappa)M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,\varphi_k)\right|^2\frac{dk_\perp\, d\phi_k}{2\pi}.\end{aligned} \tag{23}$$

We can further simplify $\sigma^{(\mathrm{tw})}_{m_f}$ by making use of Eq. (76a) from Ref. [14] to evaluate the square of the δ function

$$|\delta(k_\perp-\varkappa)|^2 = \frac{R}{\pi}\,\delta(k_\perp-\varkappa), \tag{24}$$

and using Eq. (11), which suggests that the dependence of the amplitude $M^{(\mathrm{pl})}_{m'_f m_i}(\theta_k,\varphi_k)$ on the azimuthal angle φ_k arises solely from the factor $e^{-i(m_f-m_i)\varphi_k}$:

$$\sigma^{(\mathrm{tw})}_{m_f}(\theta_k) = \frac{1}{2l_i+1}\frac{(2\pi)^2}{k_z} \times\sum_{m_i}\left|M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,0)\right|^2\delta(\omega+E_i-E_f), \tag{25}$$

where $k_z = k\cos\theta_k$. This expression is analog to Eq. (21) derived for the absorption of a plane-wave light. The only difference here—apart from the trivial prefactor—is the matrix element $M^{(\mathrm{pl})}_{m_f m_i}(\theta_k,0)$ which depends now on the angle $\theta_k = \arctan(\varkappa/k_z)$ characterizing the ratio of transverse to longitudinal components of the photon's linear momentum.

III. RESULTS AND DISCUSSION

Using Eqs. (22) and (25) we can investigate the excitation of one-electron ions by the twisted light. These expressions have been derived for two different scenarios in which the Bessel photon beam interacts with either (i) a well-localized single atom or (ii) a macroscopic atomic target. Below we shall focus on the second scenario which can be more easily realized experimentally. The partial cross section (25), derived for this scenario, is independent of the projection of the total angular momentum m_γ and sensitive only to the transverse momentum $\varkappa$ of the twisted wave as characterized by the angle θ_k. In Fig. 2 we display the θ_k dependence of the relative total

$$\frac{\sigma^{(\mathrm{tw})}_{\mathrm{tot}}(\theta_k)}{\sigma^{(\mathrm{pl})}_{\mathrm{tot}}} \equiv \frac{\sum_{m_f}\sigma^{(\mathrm{tw})}_{m_f}(\theta_k)}{\sum_{m_f}\sigma^{(\mathrm{pl})}_{m_f}}, \tag{26}$$

as well as relative partial cross sections

$$\frac{\sigma^{(\mathrm{tw})}_{m_f}(\theta_k)}{\sigma^{(\mathrm{tw})}_{\mathrm{tot}}(\theta_k)} \equiv \frac{\sigma^{(\mathrm{tw})}_{m_f}(\theta_k)}{\sum_{m_f}\sigma^{(\mathrm{tw})}_{m_f}(\theta_k)}, \tag{27}$$

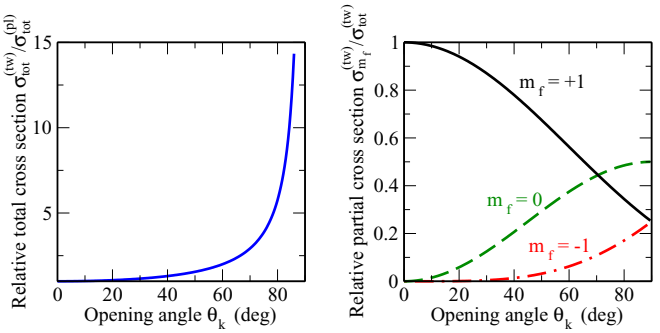

FIG. 2. (Color online) The relative total (26) and partial cross sections (27) for the $1s \to 2p$ excitation of neutral hydrogen atoms by the twisted photons. Results are presented as a function of the opening angle θ_k of the Bessel beam.

calculated for the $1s \to 2p$ excitation of neutral hydrogen by the incident photons with helicity $\lambda = +1$. As seen from the figure, the predictions obtained for the twisted light match closely with the "plane-wave" results at vanishing values of the angle θ_k. In particular, only the partial cross section $\sigma_{m_f=+1}^{(tw)}$ is *nonzero* for $\theta_k = 0$, following the selection rule (9) in which $m_i = 0$ and, hence, $m_f = \lambda = +1$. Moreover, as can be expected from Eqs. (21) and (25), the $\sigma_{m_f=+1}^{(tw)}(\theta_k = 0)$ coincides with the cross section $\sigma_{m_f=+1}^{(pl)}$, obtained for the incident plane-wave radiation.

With the increase of the transverse momentum $\varkappa$, the absorption of the twisted light may lead to the population of other substates with $m_f \neq 1$. As seen from the right panel of Fig. 2, for example, the partial cross section for the excitation to the level with $m_f = 0$ grows up with the angle θ_k and even becomes larger than $\sigma_{m_f=+1}^{tw}(\theta_k)$ for $\theta_k \gtrsim 71^\circ$. One can also observe the enhancement of the $\sigma_{m_f=-1}^{tw}(\theta_k)$ within the strongly nonparaxial regime, where $\varkappa/k_z >> 1$. To explain such a behavior we recall that the twisted (Bessel) state can be *seen* as a coherent superposition of plane waves lying on a momentum cone surface with a polar opening angle θ_k, cf. Eqs. (15) and (16) and Fig. 1. For each plane-wave component $e^{i\boldsymbol{kr}}$ and the $1s \to 2p$ transition, the standard selection rule $m'_f = \lambda = 1$ holds in which the projection m'_f is defined, however, with respect to the unit vector $\hat{\boldsymbol{k}} = \boldsymbol{k}/k = (\sin\theta_k \cos\phi_k, \sin\theta_k \sin\phi_k, \cos\theta_k)$. To study the sublevel population along the overall quantization (z) axis, a *rotation* of the atomic system by an angle θ_k is performed and results in the breakdown of the relation (9). A similar violation of the plane-wave selection rule was predicted by Afanasev and co-workers in Ref. [9] for the scenario where the position of a target atom within the wave front is specified. In that study it was shown that, depending on the impact parameter b, the p substates with the magnetic quantum numbers $m_f = 0$ and -1 can be populated under the absorption of a twisted light with the helicity $\lambda = +1$.

The θ_k behavior of the excitation cross sections $\sigma_{m_f}^{(tw)}(\theta_k)$, displayed in Fig. 2, can be also understood based on Eqs. (13), (21), and (25). By using these expressions and making some trivial algebra we find

$$\sigma_{m_f}^{(tw)}(\theta_k) = \frac{\left|d_{m_f\lambda}^{l_f}(\theta_k)\right|^2}{\cos\theta_k}\,\sigma_\lambda^{(pl)}. \tag{28}$$

For the $1s \to 2p$ transition and the helicity of the incident light $\lambda = +1$, this equation is further simplified to

$$\begin{aligned} \frac{\sigma_{m_f=0}^{(tw)}(\theta_k)}{\sigma_{tot}^{(tw)}(\theta_k)} &= \frac{\sin^2\theta_k}{2}, \\ \frac{\sigma_{m_f=\pm1}^{(tw)}(\theta_k)}{\sigma_{tot}^{(tw)}(\theta_k)} &= \frac{(1 \pm \cos\theta_k)^2}{4}, \end{aligned} \tag{29}$$

thus confirming the numerical results from Fig. 2. The total photoexcitation cross section is also readily derived from Eq. (28) and unitarity properties of the Wigner matrices:

$$\sigma_{tot}^{(tw)}(\theta_k) = \sum_{m_f} \sigma_{m_f}^{(tw)}(\theta_k) = \frac{1}{\cos\theta_k}\,\sigma_\lambda^{(pl)}. \tag{30}$$

This formula indicates that while for $\theta_k = 0$ the $\sigma_{tot}^{(tw)}(\theta_k)$ coincides with the plane-wave result, it significantly increases with the opening angle, an effect which can be observed also in the left panel of Fig. 2.

Until now we have discussed the population of atomic sublevels following the $1s \to 2p$ transition. However, the theoretical approach, developed in the previous section is general and can be used to describe the photoexcitation from *any* atomic state. In Fig. 3, for example, we display the relative total (26) and partial cross sections (27) for the $2p \to 3d$ case. Similar to before, results were obtained as a function of the opening angle θ_k of incident Bessel photons with the helicity $\lambda = +1$. As one can expect, our predictions for $\theta_k = 0$ reproduce those obtained for the plane-wave radiation. In particular, following Eq. (9) only sublevels $|3d\, m_f\rangle$ with $m_f = 0, +1, +2$ can be populated during the excitation of the *unpolarized* $2p$ state. This plane-wave selection rule is violated with the increase of the angle θ_k and, correspondingly, of the cross sections $\sigma_{m_f=-1}^{(tw)}(\theta_k)$ and $\sigma_{m_f=-2}^{(tw)}(\theta_k)$. Such a θ_k behavior of the partial cross sections can be analyzed analytically also for the $2p \to 3d$ transition. For the sake of brevity, however, we will not present here the explicit expressions for the $\sigma_{m_f}^{(tw)}(\theta_k)$ and just refer the reader to Eqs. (11) and (25).

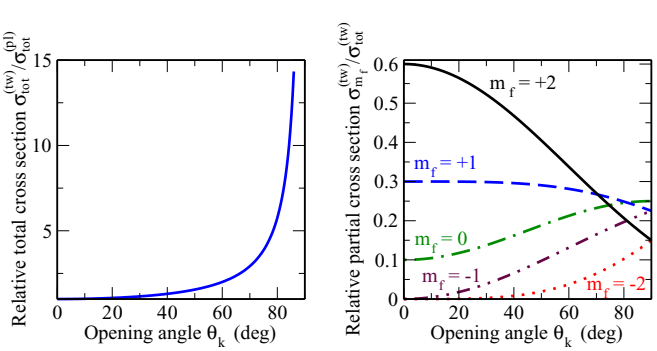

FIG. 3. (Color online) Same as Fig. 2, but for the $2p \to 3d$ transition in neutral hydrogen.

As seen from Figs. 2 and 3, the partial cross sections $\sigma^{(\mathrm{tw})}_{m_f}(\theta_k)$ for the photoinduced transitions are generally different from each other, thus leading to the *unequal* population of the final sublevels $|n_f\, l_f\, m_f\rangle$. In this case, the residual (excited) atom is said to be aligned and/or oriented [15,16]. In atomic theory, such an alignment is usually described in terms of one (or several) parameters $\mathcal{A}_{k0}$, which are related to partial excitation cross sections and can be directly employed to analyze the subsequent decay of an atom. For example, the linear polarization and angular distribution of the Lyman-α ($2p \to 1s$) radiation following the formation of the excited $2p$ state are defined by a single alignment parameter [17]:

$$\mathcal{A}_{20} = \frac{1}{2}\frac{\sigma^{(\mathrm{tw})}_{+1}(\theta_k) + \sigma^{(\mathrm{tw})}_{-1}(\theta_k) - 2\sigma^{(\mathrm{tw})}_{0}(\theta_k)}{\sigma^{(\mathrm{tw})}_{+1}(\theta_k) + \sigma^{(\mathrm{tw})}_{-1}(\theta_k) + \sigma^{(\mathrm{tw})}_{0}(\theta_k)}. \qquad (31)$$

By making use of Eqs. (29), we find that the alignment also depends on the opening angle of the twisted Bessel beam as $\mathcal{A}_{20}(\theta_k) = (1 + 3\cos 2\theta_k)/8$. The information about the θ_k can be experimentally obtained, therefore, from the analysis of the properties of the subsequent Lyman-α decay. For example, the linear polarization of the fluorescent photons, detected under the right angle with respect to the collision (z) axis, is usually characterized by the Stokes parameter P_1, which reads as [18–20]

$$P_1(\theta_k) = \frac{3\mathcal{A}_{20}(\theta_k)}{\mathcal{A}_{20}(\theta_k) - 6} = \frac{3 + 9\cos 2\theta_k}{-47 + 3\cos 2\theta_k}, \qquad (32)$$

if we assume that the fine structure of the $2p$ level remains unresolved. In experiment, this parameter is determined simply as $P_1 = (I_{||} - I_\perp)/(I_{||} + I_\perp)$, where $I_{||}$ and $I_\perp$ are the intensities of light, linearly polarized in parallel or perpendicular, respectively, to the *reaction* plane. Such a plane is spanned by the momenta $\boldsymbol{k}_z$ and $\boldsymbol{k}_{Ly-\alpha}$ of the incident Bessel and emitted fluorescent photons, respectively.

In Fig. 4 we display the polarization (32) of the Lyman-α photons emitted after the $1s \to 2p$ excitation of neutral hydrogen atoms by twisted light. Here, one can observe that the parameter $P_1(\theta_k)$ changes *qualitatively* with the opening angle θ_k. Within the paraxial regime, where $\theta_k \lesssim 5^\circ$, for example, $P_1(\theta_k)$ is relatively large and *negative* thus implying a remarkable linear polarization of the Lyman-α line in the direction perpendicular to the reaction plane. With the increase of the opening angle, $P_1(\theta_k)$ first vanishes at $\theta_k \approx 58^\circ$ and later becomes *positive* which indicates that the fluorescence emission is now predominantly polarized within the plane. Such a θ_k variation of the polarization parameter (32) can be easily observed experimentally and may provide valuable information about the interaction of twisted photon beams with atomic ensembles.

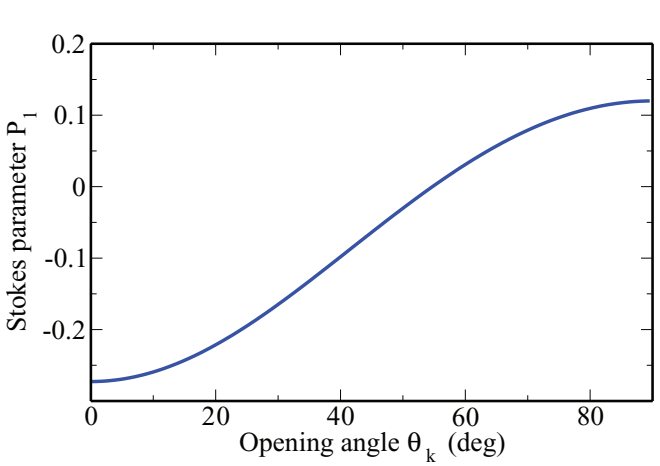

FIG. 4. (Color online) The Stokes parameter (32) of the Lyman-α radiation following the $1s \to 2p$ excitation of hydrogen atoms by twisted light with the opening angle θ_k.

As seen from Figs. 2–4 as well as the discussion above, the effect of the absorption of twisted light on the population of excited atomic states and on the linear polarization of the characteristic radiation becomes most pronounced if θ_k approaches 90°. This (right) opening angle cannot be reached experimentally and leads, moreover, to an *unphysical* situation when the radiation does not propagate along the z axis, i.e., $k_z = 0$. However, a clear distinction between the plane-wave and twisted calculations can be found also for smaller θ_k in the range from about 20° to 60°. The photoabsorption measurements employing twisted photon beams with such opening angles are likely to become feasible in the near future [21].

IV. SUMMARY AND OUTLOOK

In summary, we have performed a theoretical study of the excitation of hydrogenlike atoms by incident twisted light. Special attention was paid to the *partial* (excitation) cross sections which characterize the population of sublevels $|n_f\, l_f\, m_f\rangle$ of residual atoms. To evaluate these cross sections we employed the eigensolutions of the Schrödinger equation and the first-order perturbative approach. Based on such a nonrelativistic approach, simple analytical expressions have been derived that allow the analysis of the photoexcitation process for the experimentally realistic scenario where the twisted photon beam collides with a macroscopic atomic target. We have shown, in particular, that the partial cross sections are very sensitive to the ratio of transverse $\varkappa$ to longitudinal k_z momenta of the beam while remaining unaffected by the projection of its total angular momentum $J_z = m_\gamma$. In fact, such an insensitivity to the m_γ holds for the case when the atomic target is much larger than (the transverse extension of the) incident photon beam. In contrast, if the size of the target is small, $\lesssim 1/\varkappa$, the photoexcitation cross sections will be sensitive to the projection of the TAM. The study of this effect is out of the scope of this paper and will be presented elsewhere.

While the developed approach can be applied to investigate the photoinduced transitions between two arbitrary one-electron levels, detailed analysis has been performed for the $1s \to 2p$ and $2p \to 3d$ excitations of neutral hydrogen. Our results confirm that the standard selection rule (9), which defines the population of the (excited) substates in the course of the absorption of the plane-wave radiation, does not hold anymore for the twisted case. For example, a single transition $|1s\rangle + \gamma \to |2p\, m_f = +1\rangle$ can be induced in a hydrogen atom by the plane-wave photons with the helicity $\lambda = +1$. If, in contrast, the light is prepared in the twisted (Bessel) state, a remarkable excitation to the sublevels with $m_f \neq \lambda$ can

be observed, and the effect becomes more pronounced with the increase of the transverse momentum $\varkappa$ of the incident beam. We argued, moreover, that kinematic properties of the Bessel radiation affect not only the sublevel population of excited atomic states but also the angular distribution and linear polarization of the fluorescence emission. Experimental study of the subsequent radiative decay is feasible today and can be used to gain more insights into the fundamental light-matter interaction process with twisted particles.

Our present study was restricted to the photoexcitation of light one-electron ions for which, moreover, we neglected the effects of the spin-orbit interaction. Future experimental studies, however, most likely will focus on the interaction of twisted photons with many-electron atoms. Theoretical analysis of the bound-state transitions, induced in the complex atoms, requires accurate treatment of the relativistic and interelectronic-interaction effects. Based on the multiconfiguration Dirac-Fock method and the density matrix approach, such an analysis is currently underway and will be reported in an upcoming publication.

ACKNOWLEDGMENTS

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[1] G. Molina-Terriza, J. P. Torres, and L. Torner, Nat. Phys. **3**, 305 (2007).
[2] A. M. Yao and M. J. Padgett, Adv. Opt. Photon. **3**, 161 (2011).
[3] J. Bahrdt, K. Holldack, P. Kuske, R. Müller, M. Scheer, and P. Schmid, Phys. Rev. Lett. **111**, 034801 (2013).
[4] A. Picòn, A. Benseny, J. Mompart, J. R. Vózquez de Aldana, L. Plaja, G. F. Calvo, and L. Roso, New J. Phys. **12**, 083053 (2010).
[5] U. D. Jentschura and V. G. Serbo, Phys. Rev. Lett. **106**, 013001 (2011).
[6] O. Matula, A. G. Hayrapetyan, V. G. Serbo, A. Surzhykov, and S. Fritzsche, J. Phys. B **46**, 2050002 (2013).
[7] B. S. Davis, L. Kaplan, and J. H. McGuire, J. Opt. **15**, 035403 (2013).
[8] A. Afanasev, C. E. Carlson, and A. Mukherjee, Int. J. Mod. Phys.: Conf. Ser. **25**, 1460048 (2014).
[9] A. Afanasev, C. E. Carlson, and A. Mukherjee, Phys. Rev. A **88**, 033841 (2013).
[10] A. Afanasev, C. E. Carlson, and A. Mukherjee, arXiv:1401.1227.
[11] D. A. Varshalovich, A. N. Moskalev, and V. K. Khersonskii, *Quantum Theory of Angular Momentum* (World Scientific, Singapore, 1988).
[12] A. Surzhykov, S. Fritzsche, and P. Koval, Comput. Phys. Commun. **165**, 139 (2005).
[13] S. McConnell, S. Fritzsche, and A. Surzhykov, Comput. Phys. Commun. **181**, 711 (2010).
[14] U. D. Jentschura and V. G. Serbo, Eur. Phys. J. C **71**, 1571 (2011).
[15] K. Blum, *Density Matrix Theory and Applications* (Plenum, New York, 1996).
[16] V. V. Balashov, A. N. Grum-Grzhimailo, and N. M. Kabachnik, *Polarization and Correlation Phenomena in Atomic Collisions* (Springer-Verlag, Berlin, 2000).
[17] V. V. Sizov and N. M. Kabachnik, J. Phys. B **13**, 1601 (1980).
[18] I. C. Percival and M. J. Seaton, Phil. Trans. R. Soc. A **251**, 113 (1958).
[19] L. D. Ellsworth, B. L. Doyle, U. Schiebel, and J. R. Macdonald, Phys. Rev. A **19**, 943 (1979).
[20] F. Bell, J. Phys. B **17**, L65 (1984).
[21] M. Ornigotti (private communication).