

Off-axis excitation of hydrogenlike atoms by twisted photons

Andrei Afanasev,¹ Carl E. Carlson,² and Asmita Mukherjee³

¹*Department of Physics, The George Washington University, Washington, DC 20052, USA*

²*Department of Physics, The College of William and Mary in Virginia, Williamsburg, VA 23187, USA*

³*Department of Physics, Indian Institute of Technology Bombay, Powai, Mumbai 400076, India*

(Received 17 May 2013; published 24 September 2013)

We show that the twisted-photon states, or photon states with large ($>\hbar$) angular momentum projection (m_γ) in the direction of motion, can photoexcite atomic levels for a hydrogenlike atom that are novel and distinct and are not restricted by m_γ , when the symmetry axis of the twisted-photon beam does not coincide with the center of the atomic target. Selection rules are given and interesting implications and observables for the above process are pointed out.

DOI: [10.1103/PhysRevA.88.033841](https://doi.org/10.1103/PhysRevA.88.033841)

PACS number(s): 42.50.Ct, 32.80.-t

I. INTRODUCTION AND MOTIVATION

The fact that circularly polarized photons carry an angular momentum \hbar was predicted theoretically and demonstrated experimentally in a seminal experiment by Beth in 1936 [1]. It was also realized [2] (Appendix) that the electromagnetic wave can carry orbital angular momentum in the direction of its propagation if it is constrained in the transverse plane, as in waveguides. Much later, in 1992, Allen and collaborators suggested [3] that special light beams that can propagate in vacuum, called Laguerre-Gaussian beams, predicted as non-plane-wave solutions of Maxwell equations, can carry large angular momentum $J_z \gg \hbar$ associated with their helical wave fronts. At a quantum level such beams can be described in terms of “twisted photons” [4]. This concept can be also extended to beams of particles, and electrons in particular [5]. Detailed reviews of the field are published, cf. [6]. Methods to produce “twisted” light include spiral phase plates, computer-generated holograms [6], and synchrotron radiation in a helical undulator [7,8] or in a free-electron laser [9]. Theoretical work [10,11] has shown that one can generate twisted photons with high energies of several GeV via Compton backscattering of laser photons on an energetic electron beam, making such photon beams relevant for nuclear and particle physics.

An important question is, to what extent is absorption of the twisted photons by atoms or nuclei different from the plane-wave photons? Work by Picón *et al.* [12] demonstrated that during photoionization of atoms, the knocked-out electrons carry angular momenta that reproduce the angular momentum of the incoming photons. The references [12,13] deal mainly with a special case in which the photon beam’s symmetry axis coincides with a center of an atom. In a recent publication [14] the authors analyzed elastic scattering of the twisted photons on a hydrogen atom, again with a restriction that the atom is located at the center of the optical vortex. We consider a more general case of arbitrary positioned beams and considered photoexcitation of bound states with different quantum numbers in a hydrogen atom. Such considerations can also be found in [15], which has important observations regarding total angular momentum conservation, although many formulas are given just for the atom-on-axis case. After presenting theoretical formalism for excitation of an atom by twisted photons, we point out novel effects caused by large

angular momenta of the incoming photons. Our arguments are further corroborated by theoretical calculations showing that a significant fraction of the atomic levels excited by the twisted photons could not be otherwise produced by plane-wave photons. The angular momentum projection of the twisted-photon state is derived in the Appendix.

II. BASIC FORMULAS

The twisted-photon definition here follows Serbo and Jentschura [10,11], although with a more field-theory-based viewpoint. Another possibility would be to quantize a Laguerre-Gaussian laser mode considered in the original work by Allen *et al.* [3], but our main conclusions will not be affected by this choice.

A twisted photon moving in the z direction is

$$\begin{aligned} |\kappa m_\gamma k_z \Lambda\rangle &= \int \frac{d^2 k_\perp}{(2\pi)^2} a_{\kappa m_\gamma}(\vec{k}_\perp) |\vec{k}, \Lambda\rangle \\ &= \sqrt{\frac{\kappa}{2\pi}} \int \frac{d\phi_k}{2\pi} (-i)^{m_\gamma} e^{im_\gamma \phi_k} |\vec{k}, \Lambda\rangle, \end{aligned} \quad (1)$$

where $|\vec{k}, \Lambda\rangle$ are plane-wave states, or momentum eigenstates with fixed longitudinal component k_z and fixed magnitude traverse component

$$a_{\kappa m_\gamma}(\vec{k}_\perp) = (-i)^{m_\gamma} e^{im_\gamma \phi_k} \sqrt{\frac{2\pi}{\kappa}} \delta(\kappa - |\vec{k}_\perp|). \quad (2)$$

The twisted-photon state can thus be viewed as a superposition of plane-wave states where the momenta form a cone in momentum space with a fixed pitch angle

$$\theta_k = \arctan\left(\frac{|\vec{k}_\perp|}{k_z}\right), \quad (3)$$

and varying azimuthal angle weighted by a phase $e^{im_\gamma \phi_k}$.

The normalization is

$$\begin{aligned} \langle \kappa' m'_\gamma k'_z \Lambda' | \kappa m_\gamma k_z \Lambda \rangle \\ = 2\pi 2\omega \delta(k_z - k'_z) \delta(\kappa - \kappa') \delta_{m_\gamma m'_\gamma} \delta_{\Lambda \Lambda'} \end{aligned} \quad (4)$$

for $\langle \vec{k}' \Lambda' | \vec{k} \Lambda \rangle = (2\pi)^3 2\omega \delta^3(\vec{k} - \vec{k}') \delta_{\Lambda \Lambda'}$, and $\omega = |\vec{k}|$.

Using the photon field operator $A^\mu(x)$, the wave function of a plane-wave photon is

$$\langle 0|A^\mu(x)|\vec{k}, \Lambda\rangle = \varepsilon_{\vec{k}, \Lambda}^\mu e^{-ikx} \quad (5)$$

and so the wave function of the twisted photon is

$$\begin{aligned} \mathcal{A}_{\kappa m_\gamma k_z \Lambda}^\mu(x) &= \langle 0|A^\mu(x)|\kappa m k_z \Lambda\rangle \\ &= \sqrt{\frac{\kappa}{2\pi}} \int \frac{d\phi_k}{2\pi} (-i)^{m_\gamma} e^{im_\gamma \phi_k} \varepsilon_{\vec{k}, \Lambda}^\mu e^{-ikx}. \end{aligned} \quad (6)$$

In cylindrical coordinates this is

$$\begin{aligned} \mathcal{A}_{\kappa m_\gamma k_z \Lambda}^\mu(x) &= e^{-i(\omega t - k_z z)} \\ &\times \sqrt{\frac{\kappa}{2\pi}} \int \frac{d\phi_k}{2\pi} (-i)^{m_\gamma} e^{im_\gamma \phi_k} \varepsilon_{\vec{k}, \Lambda}^\mu e^{i\vec{k}_\perp \cdot \vec{x}_\perp}, \end{aligned} \quad (7)$$

so that the twisted photon in coordinate space has a self-reproducing two-dimensional wave front moving forward at a speed less than the normal speed of light.

The wave front can be given explicitly with help of the Jacobi-Anger formula

$$e^{i\vec{k}_\perp \cdot \vec{x}_\perp} = \sum_{n=-\infty}^{\infty} i^n e^{in(\phi_\rho - \phi_k)} J_n(\kappa\rho), \quad (8)$$

where ϕ_ρ is the azimuthal angle in coordinate space, $\rho = |\vec{x}_\perp|$, and J_n is the Bessel function, and with

$$\begin{aligned} \varepsilon_{\vec{k}, \Lambda}^\mu &= e^{-i\Lambda\phi_k} \cos^2 \frac{\theta_k}{2} \eta_\Lambda^\mu + e^{i\Lambda\phi_k} \sin^2 \frac{\theta_k}{2} \eta_{-\Lambda}^\mu \\ &+ \frac{\Lambda}{\sqrt{2}} \sin \theta_k \eta_0^\mu, \end{aligned} \quad (9)$$

where the η 's are constant vectors,

$$\eta_{\pm 1}^\mu = \frac{1}{\sqrt{2}}(0, \mp 1, -i, 0), \quad \eta_0^\mu = (0, 0, 0, 1); \quad (10)$$

the photon polarization vector phase is like the Trueman-Wick [16] phase convention. Then

$$\begin{aligned} \mathcal{A}_{\kappa m_\gamma k_z \Lambda}^\mu(x) &= e^{-i(\omega t - k_z z)} \sqrt{\frac{\kappa}{2\pi}} \left\{ \frac{\Lambda}{\sqrt{2}} e^{im_\gamma \phi_\rho} \sin \theta_k J_{m_\gamma}(\kappa\rho) \eta_0^\mu \right. \\ &+ i^{-\Lambda} e^{i(m_\gamma - \Lambda)\phi_\rho} \cos^2 \frac{\theta_k}{2} J_{m_\gamma - \Lambda}(\kappa\rho) \eta_\Lambda^\mu \\ &\left. + i^\Lambda e^{i(m_\gamma + \Lambda)\phi_\rho} \sin^2 \frac{\theta_k}{2} J_{m_\gamma + \Lambda}(\kappa\rho) \eta_{-\Lambda}^\mu \right\}. \end{aligned} \quad (11)$$

As an aside, if we were to write the photon wave function for a plane-wave photon of helicity Λ , it would be like the above, possibly with some differences of normalization choice, but with pitch angle $\theta_k \rightarrow 0$ (including $\kappa \rightarrow 0$) and without an azimuthal phase factor, i.e., equivalent to $m_\gamma = \Lambda$ in the preceding equation.

The twisted-photon wave front has the feature that the Poynting vector is spiraling forward. It has azimuthal and z components in cylindrical coordinates, but no radial

component. In detail, the magnetic field for $\Lambda = 1$ is

$$\begin{aligned} B_\rho &= i\omega \sqrt{\frac{\kappa}{4\pi}} e^{i(k_z z - \omega t + m_\gamma \phi)} \\ &\times \left(\sin^2 \frac{\theta_k}{2} J_{m_\gamma + 1}(\kappa\rho) + \cos^2 \frac{\theta_k}{2} J_{m_\gamma - 1}(\kappa\rho) \right), \\ B_\phi &= \omega \sqrt{\frac{\kappa}{4\pi}} e^{i(k_z z - \omega t + m_\gamma \phi)} \\ &\times \left(\sin^2 \frac{\theta_k}{2} J_{m_\gamma + 1}(\kappa\rho) - \cos^2 \frac{\theta_k}{2} J_{m_\gamma - 1}(\kappa\rho) \right), \\ B_z &= \omega \sqrt{\frac{\kappa}{4\pi}} e^{i(k_z z - \omega t + m_\gamma \phi)} \sin \theta_k J_{m_\gamma}(\kappa\rho), \end{aligned} \quad (12)$$

and the electric field is just 90° out of phase with the magnetic field, $\vec{E} = i\vec{B}$. The physical fields are the real parts of the above expressions, and one can see that the wave front moves forward at less than the normal speed of light. Working physical electric and magnetic fields, the Poynting vector $\vec{S} = \vec{E} \times \vec{B}$ is

$$\begin{aligned} S_\rho &= 0, \\ S_\phi &= \frac{\kappa\omega^2}{4\pi} \sin \theta_k J_{m_\gamma}(\kappa\rho) \\ &\times \left(\cos^2 \frac{\theta_k}{2} J_{m_\gamma - 1}(\kappa\rho) + \sin^2 \frac{\theta_k}{2} J_{m_\gamma + 1}(\kappa\rho) \right), \\ S_z &= \frac{\kappa\omega^2}{4\pi} \left(\cos^4 \frac{\theta_k}{2} J_{m_\gamma - 1}^2(\kappa\rho) - \sin^4 \frac{\theta_k}{2} J_{m_\gamma + 1}^2(\kappa\rho) \right). \end{aligned} \quad (13)$$

Figure 1 shows S_ϕ in the transverse plane. For this illustration, and for the next, the photon wavelength is 0.5 microns, the pitch angle is 0.2 radians, and $m_\gamma = 4$. The figure shows a bulls-eye pattern characteristic of twisted photons, with a wide hole in the middle that one can also see from the Bessel functions in the above expressions. Figure 2 shows \vec{S} in the transverse plane, showing again the bulls-eye pattern and also showing the circulation of momentum density about the center of the pattern.

The center of the bulls-eye is currently at the origin in the x - y plane. Shifting it is easily done by applying the translation operator $\exp\{i\hat{p}\cdot b\}$ to the twisted-photon state $|\kappa m_\gamma k_z \Lambda\rangle$, where \hat{p}^μ is the momentum operator and b^μ is a constant vector. There is a *de facto* phase convention in Eq. (5) that the momentum eigenstate wave function at $x = 0$ is just the polarization vector. Algebraic effects of the shift are to change

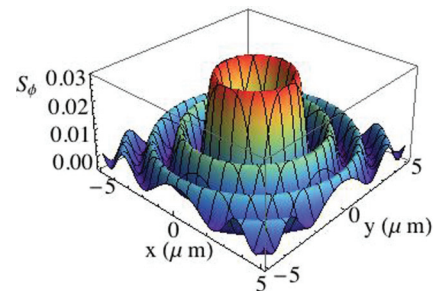


FIG. 1. (Color online) The size of Poynting vector azimuthal component as a function of position in the transverse plane. For this illustration, the photon wavelength is 0.5 microns, the pitch angle is 0.2 radians, and $m_\gamma = 4$.

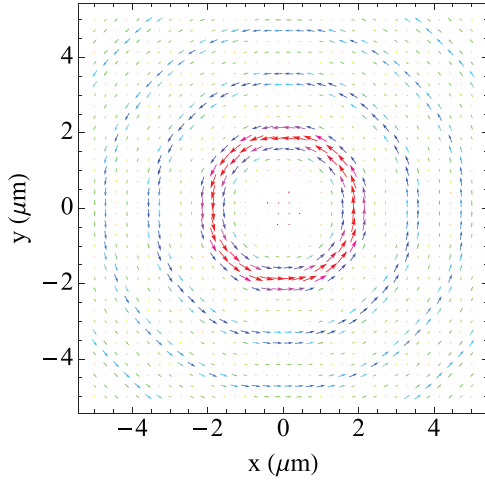


FIG. 2. (Color online) $2\pi\rho \times \vec{S}$ projected onto the transverse plane. One sees the major bands of \vec{S} circulating in the same direction, building up the large orbital angular momentum. Also for this illustration, $\lambda = 0.5 \mu\text{m}$, $\theta_k = 0.2$ radians, and $m_\gamma = 4$.

the phase in Eq. (6) from $\exp\{-ikx\}$ to $\exp\{-ik(x-b)\}$ and arguments of the Bessel functions in later equations to $J_\nu(\kappa|\vec{x}_\perp - \vec{b}_\perp|)$.

III. ATOMIC PHOTOEXCITATION

We will consider excitation by a twisted photon of a hydrogenlike atom from the ground state to an excited state. In general, the photon's wave front will be traveling in the z

direction and the axis of the twisted photon will be displaced from the nucleus of the atomic target by some distance in the x - y plane which we will call \vec{b} . We work out the photoexcitation for this case in this section and then apply the result to two situations. One situation will be the case when the twisted-photon axis passes directly through the center of the atom's nucleus. The other will be the case where target atoms are at random locations, and we have to average over all axis-to-atom separations.

For simplicity, we will treat an atomic state just in terms of its principal quantum number n_k , orbital quantum number l_k , and magnetic quantum number m_k , where $k = i$ for the initial state and $k = f$ for the final state. We treat the atom nonrelativistically. The twisted photon satisfies the Coulomb gauge condition, and the interaction Hamiltonian is

$$H_1 = -\frac{e}{m_e} \vec{A} \cdot \vec{p}, \quad (14)$$

and the transition matrix element is

$$S_{fi} = -i \int dt \langle n_f l_f m_f | H_1 | n_i l_i m_i; \kappa m k_z \Lambda \rangle. \quad (15)$$

The twisted-photon wave function is given in Eq. (11).

We shall center the atomic nucleus at the origin, with the atomic electron located at (ρ, ϕ_ρ, z) in cylindrical coordinates or (r, θ_r, ϕ_ρ) in spherical coordinates. The twisted photon, moving in the z direction, has its origin in general not centered on the atomic nucleus but displaced to position \vec{b} in the x - y plane. Relative to the photon axis, the electron position projected onto the x - y plane will be at distance $|\vec{\rho} - \vec{b}|$ and angle ϕ'_ρ , as illustrated in Fig. 3. Then

$$S_{fi} = 2\pi i \delta(E_f - E_i - \omega) \frac{e}{m_e} \sqrt{\frac{\kappa}{2\pi}} \int r^2 dr d(\cos \theta_r) d\phi_\rho R_{n_f l_f}(r) Y_{l_f m_f}^*(\theta_r, 0) e^{-im_f \phi_\rho} \left\{ \frac{\Lambda}{\sqrt{2}} e^{im_\gamma \phi'_\rho} \sin \theta_k J_{m_\gamma}(\kappa|\vec{\rho} - \vec{b}|) \vec{\eta}_0 \right. \\ \left. + i^{-\Lambda} e^{i(m_\gamma - \Lambda)\phi'_\rho} \cos^2 \frac{\theta_k}{2} J_{m_\gamma - \Lambda}(\kappa|\vec{\rho} - \vec{b}|) \vec{\eta}_\Lambda + i^\Lambda e^{i(m_\gamma + \Lambda)\phi'_\rho} \sin^2 \frac{\theta_k}{2} J_{m_\gamma + \Lambda}(\kappa|\vec{\rho} - \vec{b}|) \vec{\eta}_{-\Lambda} \right\} e^{ik_z z} \cdot \vec{p} R_{10}(r) Y_{00}, \quad (16)$$

where $E_k = E_{n_k}$. Note that

$$\hat{\eta}_\lambda \cdot \vec{p} R_{10}(r) = -i \hat{\eta}_\lambda \cdot \hat{r} R'_{10}(r) = -i \sqrt{\frac{4\pi}{3}} Y_{1\lambda}(\theta_r, \phi_\rho) R'_{10}(r). \quad (17)$$

The expansion theorem,

$$e^{in\phi'_\rho} J_n(\kappa|\vec{\rho} - \vec{b}|) = \sum_{N_1=-\infty}^{\infty} e^{iN_1\phi_\rho} e^{-i(N_1-n)\phi_b} J_{N_1}(\kappa\rho) J_{N_1-n}(\kappa b), \quad (18)$$

allows us to perform the $d\phi_\rho$ integral and obtain

$$S_{fi} = -2\pi \delta(E_f - E_i - \omega) \frac{e}{m_e a_0} \sqrt{\frac{2\pi\kappa}{3}} e^{i(m_\gamma - m_f)\phi_b} J_{m_f - m_\gamma}(\kappa b) \\ \times i^{-\Lambda} \left\{ \cos^2 \frac{\theta_k}{2} g_{n_f l_f m_f \Lambda} + \frac{i}{\sqrt{2}} \sin \theta_k g_{n_f l_f m_f 0} - \sin^2 \frac{\theta_k}{2} g_{n_f l_f m_f, -\Lambda} \right\} \\ \stackrel{\text{def}}{=} 2\pi \delta(E_f - E_i - \omega) \mathcal{M}_{n_f l_f m_f \Lambda}(b). \quad (19)$$

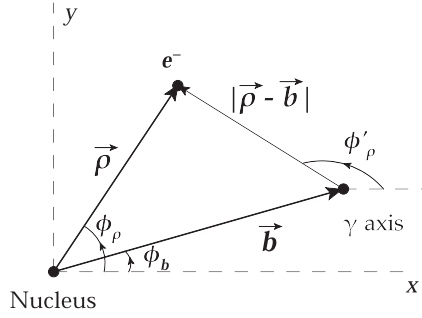


FIG. 3. Relative positions of atomic state and photon axis, as projected onto the x - y plane, with the origin at the nucleus of the atom.

Note that the energy δ function requires energy conservation, but in our formalism atomic recoil is neglected, so that overall linear momentum is not conserved. The dimensionless atomic factors are

$$\begin{aligned} g_{n_f l_f m_f \lambda} &\equiv -a_0 \int_0^\infty r^2 dr R_{n_f l_f}(r) R'_{10}(r) \\ &\times \int_{-1}^1 d(\cos \theta_r) J_{m_f - \lambda}(\kappa \rho) Y_{l_f m_f}(\theta_r, 0) Y_{1\lambda}(\theta_r, 0) e^{ik_z z}, \end{aligned} \quad (20)$$

and a_0 is the Bohr radius. The quantum numbers of the initial state are tacit, as we always start from the ground state. As a simple practical matter, $-a_0 R'_{10}(r) = R_{10}(r)$, and one also has $\kappa \rho = \omega r \sin \theta_r \sin \theta_k$ and $k_z z = \omega r \cos \theta_r \cos \theta_k$. Further, one can show that the three terms in the curly bracket above are either all real or all purely imaginary.

IV. ON-AXIS AND OFF-AXIS ATOMIC EXCITATION

By way of review, the selection rules for photoexcitation (starting from the ground state) with plane-wave photons of helicity Λ are, cf. Refs. [17],

$$m_f = \Lambda, \quad l_f \geq 1, \quad g_{n_f l_f m_f \Lambda}^{(\text{pw})} \propto (\omega a_0)^{l_f - 1}, \quad (21)$$

where $g^{(\text{pw})}$ is the plane-wave analog of the reduced atomic amplitudes shown in Eq. (20) and shows the suppression that follows when higher photon partial waves are needed.

From Eq. (19) it can be seen that the magnitude of the result depends on m_γ only through the argument of the Bessel function $J_{m_f - m_\gamma}(\kappa b)$. When a twisted photon strikes an atom centered on its axis, the impact parameter $b = 0$, and we immediately obtain $m_f = m_\gamma$ from the Bessel function in the general result, Eq. (19),

$$J_{m_f - m_\gamma}(\kappa b) \rightarrow J_{m_f - m_\gamma}(0) = \delta_{m_f m_\gamma}. \quad (22)$$

That is, the only final states that can be produced are those that can absorb the full projected angular momentum of the twisted photon.

However, the atom does not have to be far off the photon axis before other amplitudes, not satisfying the above selection rule, play an important role. This was also observed in the simulations done in [13]. In our numerical calculations as well

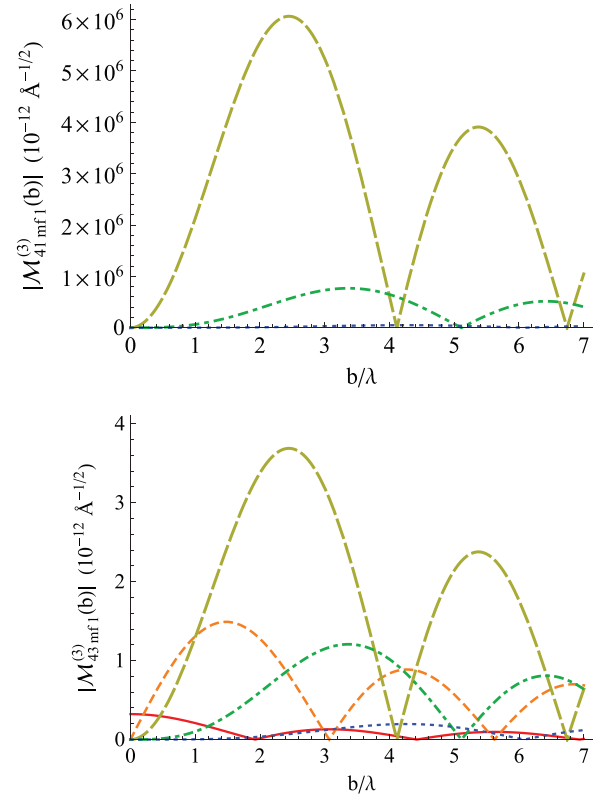


FIG. 4. (Color online) Size of the transition amplitude $|\mathcal{M}_{n_f l_f m_f \Lambda}^{(m_\gamma=3)}(b)|$ for particular quantum numbers n_f, l_f, Λ and several m_f ; here the photons are circularly polarized. Upper graph is for the final state $n_f = 4, l_f = 1$; the state $m_f = 1$ is allowed by electric-dipole selection rules for plane waves, while $m_f = 0, -1$ are unique for the twisted photons. Lower graph is for the final state $n_f = 4, l_f = 3$. The curve styles for both graphs: $m_f = 3$ is the red solid curve, $m_f = 2$ is orange and medium dashed, $m_f = 1$ is gold and long dashed, $m_f = 0$ is green and dot-dashed, $m_f = -1$ is blue and dotted, and transitions to other m_f are quite small and not plotted.

as in the plots, unless otherwise stated, we take a twisted-photon state of wavelength 97.2 nm (set by the hydrogen atom spacing), $m_\gamma = 3$, $\Lambda = 1$, and $\theta_k = 0.2$ radians. As an illustration, we plot in Fig. 4 the amplitudes $|\mathcal{M}_{n_f l_f m_f \Lambda}(b)|$ for the example of $n_f = 4, l_f = 1$ and photon angular momentum along the direction of motion $m_\gamma = 3$ (upper plot) and $n_f = 4, l_f = 3$ (lower plot). Note the relative strength of the amplitudes is much higher, by about six orders of magnitudes, for the transition into $l_f = 1$ state vs $l_f = 3$, in accordance with the selection rules presented below.

In Fig. 4 the horizontal axis is the impact parameter b in units of the photon wavelength λ . Already with an impact parameter of less than half a wavelength, amplitudes that do not satisfy the $m_f = m_\gamma$ selection rule are becoming important. The amplitude with the largest peak is the one with $m_f = 1$, which is the only amplitude one would have with a plane-wave photon polarized with helicity $\Lambda = 1$. In a near-field setup, when the twisted light outside of the central (small- b) areas is blocked by a screen, one can study whether the enhancement of the transition to $m_f = m_\gamma$ for the photons with large m_γ , as seen in Fig. 4 (lower plot, continuous curve), would lead

to enhanced attenuation of these photons by atoms, making matter more opaque to them.

The general selection rules for the off-axis case are

$$m_f = \text{any}, \quad l_f \geq |m_f|, \\ g_{n_f l_f, m_f, \Lambda} \propto (\omega a_0 \cos \theta_k)^{|l_f - |m_f - \Lambda|| - 1} (\omega a_0 \sin \theta_k)^{|m_f - \Lambda|}, \quad (23)$$

where in the last line, the first factor is absent if its exponent is negative. Note that we neglect atomic recoil. For manifest conservation of both linear and angular momentum, the atom's recoil momentum can be taken into account [15].

V. CROSS SECTIONS FOR RANDOMLY DISTRIBUTED TARGETS

A. Cross-section calculation

In general, the location of the photon axis cannot be controlled at the level of the photon wavelength, so we should average over the transverse separations. For transverse separation \vec{b} ,

$$\sigma_{n_f l_f m_f \Lambda}^{(m_\gamma)} = 2\pi \delta(E_f - E_i - \omega_\gamma) \frac{|\mathcal{M}_{n_f l_f m_f \Lambda}^{(m_\gamma)}(b)|^2}{f}, \quad (24)$$

where f is the incoming flux and a suitable sum or average over spins is implied.

With the target atoms uniformly distributed, the cross section averaged over atom location is

$$\begin{aligned} \bar{\sigma}_{n_f l_f m_f \Lambda} &= \frac{2\pi \delta(E_f - E_i - \omega)}{f} \frac{1}{\pi R^2} \int d^2b |\mathcal{M}_{n_f l_f m_f \Lambda}(b)|^2 \\ &= \frac{2\pi \delta(E_f - E_i - \omega)}{f} \frac{1}{\pi R^2} \frac{2\pi e^2 \kappa}{3m_e^2 a_0^2} \\ &\quad \times \int_0^R 2\pi b db J_{m_f - m_\gamma}^2(\kappa b) \left| \cos^2 \frac{\theta_k}{2} g_{n_f l_f m_f \Lambda} \right. \\ &\quad \left. + \frac{i}{\sqrt{2}} \sin \theta_k g_{n_f l_f m_f 0} - \sin^2 \frac{\theta_k}{2} g_{n_f l_f m_f, -\Lambda} \right|^2. \end{aligned} \quad (25)$$

Outside the integration measure, the only b dependence is in the Bessel function, and the useful integral is

$$\lim_{R \rightarrow \infty} \int_0^R b db J_{m_f - m_\gamma}^2(\kappa b) = \frac{R}{\pi \kappa}, \quad (26)$$

independent of index.

For the flux we take the average density of the twisted-photon state times the incoming wave front velocity k_z/ω . The target is unit normalized. The density of the twisted-photon state, with our normalization and averaged over a disk of radius R , can be worked out and leads to

$$f = \rho_{\text{avg}} \frac{k_z}{\omega} = \frac{2k_z}{\pi^2 R}. \quad (27)$$

Thus

$$\begin{aligned} \bar{\sigma}_{n_f l_f m_f \Lambda} &= 2\pi \delta(E_f - E_i - \omega) \frac{8\pi^3 \alpha^3}{3k_z} \left| \cos^2 \frac{\theta_k}{2} g_{n_f l_f m_f \Lambda} \right. \\ &\quad \left. + \frac{i}{\sqrt{2}} \sin \theta_k g_{n_f l_f m_f 0} - \sin^2 \frac{\theta_k}{2} g_{n_f l_f m_f, -\Lambda} \right|^2. \end{aligned} \quad (28)$$

B. Unique twisted-photon features

For the twisted photon centered on target, there is the dramatic result that the magnetic quantum number of the final atomic state must equal the corresponding z projection of the angular momentum of the twisted photon. This constraint is relaxed for the general case of random target location, but there are still features unique to twisted photons.

Photoexcitation, starting from the ground state, by a plane-wave photon of a certain helicity leads only to final states whose magnetic quantum number equals the helicity. Twisted photons, on the other hand, photoexcite states with a large range of magnetic quantum numbers m_f . Values of m_f impossible for plane-wave photons are produced even when the twisted photons enter a medium with random target locations.

Twisted photons also produce the $m_f = \Lambda$ states that plane-wave photons necessarily lead to. But the interest is in the $m_f \neq \Lambda$ states unique to twisted-photon production. To quantify the probability of finding these states, we define a ratio for a fixed Λ which compares the rate for producing final states that are unique to twisted photons to the total rate where the twisted photon produces all final states, including $m_f = \Lambda$, for a given energy level characterized by quantum numbers (n_f, l_f) (and for the case of a large interaction region with random target locations),

$$f_{\text{twisted}} = \frac{\sum_{\substack{m_f = l_f \\ m_f \neq \Lambda}} \bar{\sigma}_{n_f l_f m_f \Lambda}}{\sum_{m_f = -l_f}^{m_f = l_f} \bar{\sigma}_{n_f l_f m_f \Lambda}}. \quad (29)$$

The twisted-photon ratio f_{twisted} evaluates what fraction of the final states excited by the twisted photon could not have been produced by a plane-wave photon. As a numerical example, we evaluate this ratio for final states with varied values of n_f and l_f and the result is

$$\begin{aligned} f_{\text{twisted}}[\text{g.s.} \rightarrow (n_f = 4, l_f = 1)] &= 2.0\%, \\ f_{\text{twisted}}[\text{g.s.} \rightarrow (n_f = 4, l_f = 3)] &= 20.3\%. \end{aligned} \quad (30)$$

A comparison between the total photoproduction rate from twisted photons and from plane-wave photons is

$$r_{\text{twisted}} = \frac{\sum_{m_f = -l_f}^{m_f = l_f} \bar{\sigma}_{n_f l_f m_f \Lambda}}{\sigma_{n_f l_f \Lambda \Lambda}^{(\text{pw})}}. \quad (31)$$

For the above-chosen final states, the other ratio works out to $r_{\text{twisted}} = 1.02$.

The ratios f_{twisted} and r_{twisted} can be measured in experiments. They can provide a tool to identify twisted photons arriving from pointlike sources, no matter if they come from distant stars or are produced in the laboratory. In particular, the presence of the $m_f = 0$ state in the atomic excitation produced by the photons coming from a well-defined direction would be a definitive signal of a twisted-photon absorption.

Let us discuss one more special feature of the twisted photons and compare the probabilities of photoexcitation of an (unpolarized) atom by the photons with opposite helicities $\pm \Lambda$. For plane-wave photons these probabilities are identical due to parity conservation. For twisted photons with a fixed z projection of orbital angular momentum but opposite helicities

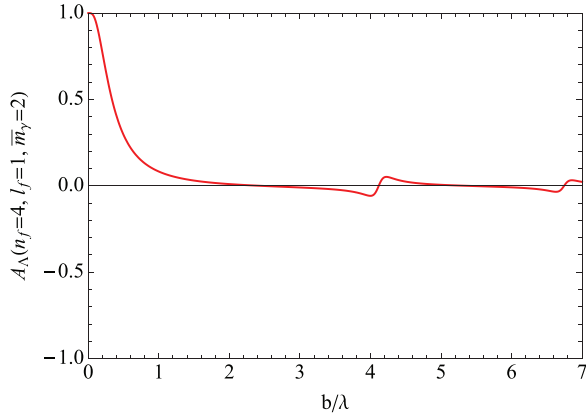


FIG. 5. (Color online) Helicity asymmetry A_Λ for excitation of the atomic state $n_f = 4, l_f = 1$ by a twisted photon with $\bar{m}_\gamma = 2$.

such an asymmetry would not violate parity, because the corresponding photon states do not transform into each other via parity transformation. The corresponding helicity asymmetry A_Λ can be defined as

$$A_\Lambda(n_f, l_f, \bar{m}_\gamma) = \frac{\sum_{m_f=l_f}^{m_f=l_f} (\sigma_{n_f l_f m_f \Lambda=-1}^{\bar{m}_\gamma-1} - \sigma_{n_f l_f m_f \Lambda=1}^{\bar{m}_\gamma+1})}{\sum_{m_f=-l_f}^{m_f=l_f} (\sigma_{n_f l_f m_f \Lambda=-1}^{\bar{m}_\gamma-1} + \sigma_{n_f l_f m_f \Lambda=1}^{\bar{m}_\gamma+1})}. \quad (32)$$

The value of \bar{m}_γ is zero for plane-wave photons; for the twisted photons it is controlled by the method of their generation, and in a paraxial approximation it would correspond to z projection of the orbital angular momentum. The asymmetry A_Λ turns to zero after averaging up to infinite values of the impact parameter b ; it is large for the central areas of the optical vortex, as shown in Fig. 5. This interesting observable can be studied experimentally for microparticles placed in the inner area of the optical vortex; it indicates that the electromagnetic fields due to spin and orbital angular momentum of the twisted photons add up coherently, leading to distinctively different strength of interaction at a given transverse-plane location if the direction of spin is flipped. The corresponding figure of merit $A_\Lambda^2 \sigma$ is small at $b = 0$, but peaks near $b/\lambda = 0.6$, where A_Λ is about 20%.

VI. SUMMARY AND DISCUSSION

In this paper we developed a formalism for photoexcitation of an atom with a beam of twisted photons, using a hydrogen atom as an example. In the derivation, we use an expansion [10,11] of the twisted-photon states in terms of plane waves. We show that in a special case when the photon beam axis coincides with the atomic center, the transitions between atomic levels obey angular momentum selection rules, similar to the conclusions made in Ref. [12] for photoionization, resulting in an excited state with a magnetic quantum number ($J_z = m_1$) exactly matching m_γ of the incoming beam. We also recover standard electric-dipole angular momentum selection rules [17,18] in the limit of plane-wave photons.

Next, we extended our calculation to a more general case of the atoms located away from the photon beam axis and analyzed the amplitudes of various transitions as a function of

the beam center position in units of photon wavelength, b/λ . In this case the magnetic quantum number of the photoexcited state no longer matches m_γ , and a range of final-state quantum numbers is generated. Relative magnitudes of various transitions were studied both analytically and numerically. It was found that after we average over the beam position, the amplitude allowed by standard plane-wave dipole selection rules quickly gains strength and makes a dominant contribution to photoabsorption.

To quantify the role of photon orbital angular momentum, we introduced several observables that describe probabilities of excitation of the $m_f \neq \Lambda$ states that are forbidden to plane waves. The important finding is that relative probability of transitions to “forbidden” states can reach tens of percent in the examples we considered ($\lambda = 100$ nm, pitch angle = 0.2 rad, $m = 3$). This is in stark contrast with familiar selection rules [17,18]. Given a noticeable effect arising from the orbital angular momentum of the photons, our predictions can be verified experimentally.

ACKNOWLEDGMENTS

C.E.C. thanks the National Science Foundation for support under Grants No. PHY-0855618 and No. PHY-1205905. Work of A.A. was supported by The George Washington University. A.M. thanks Jefferson Lab and the College of William and Mary for hospitality during the initial stages of this work.

APPENDIX: ANGULAR MOMENTUM PROJECTION

The factor $\exp\{im_\gamma \phi_k\}$ in the twisted-photon state should give the state a z component of angular momentum that is at least approximately m . This is stated in a number of sources, but we have not seen a claim of exactness. In fact we can prove that the total angular momentum projected in the longitudinal direction is exactly m_γ , at least in the sense of expectation values.

From the Noether current corresponding to rotations, one gets the angular momenta. The result can be found, for example, in Bjorken and Drell [19], Eq. (14.22), and is

$$J^{ij} = \varepsilon^{ijk} J^k \\ = \int d^3x : \vec{A} \cdot (x^i \partial^j - x^j \partial^i) \vec{A} - (\dot{A}^i A^j - \dot{A}^j A^i) : \quad (A1)$$

We will speak of the first term as the orbital angular momentum and the second as the spin.

Regarding the spin term, one can consider a direct calculation with the usual expansion and commutation relation in terms of plane-wave states,

$$A^\mu(x) = \sum_\lambda \int \frac{d^3q}{(2\pi)^3 2\omega_q} (a_{\vec{q}\lambda} \varepsilon_{\vec{q}\lambda}^\mu e^{-iqx} + a_{\vec{q}\lambda}^\dagger \varepsilon_{\vec{q}\lambda}^{\mu*} e^{iqx}), \quad (A2)$$

and

$$[a_{\vec{q}\lambda}, a_{\vec{k}\Lambda}^\dagger] = (2\pi)^3 2\omega \delta^3(\vec{q} - \vec{k}) \delta_{\lambda\Lambda}. \quad (A3)$$

After noting that the $a^\dagger a^\dagger$ and aa terms in $J^3(\text{spin})$ do not contribute to the matrix element below, one can show the $a^\dagger a$

terms lead to

$$\langle k' \Lambda' | J^3(\text{spin}) | k \Lambda \rangle = 2i\omega(2\pi)^3 \delta^3(\vec{k} - \vec{k}') (\vec{\varepsilon}_{\vec{k}\Lambda}^- \times \vec{\varepsilon}_{\vec{k}'\Lambda'}^*)^z. \quad (\text{A4})$$

After showing

$$(\vec{\varepsilon}_{\vec{k}\Lambda}^- \times \vec{\varepsilon}_{\vec{k}'\Lambda'}^*)^z = -i\Lambda \cos \theta_k \delta_{\Lambda\Lambda'}, \quad (\text{A5})$$

this becomes

$$\langle k' \Lambda' | J^3(\text{spin}) | k \Lambda \rangle = \Lambda \cos \theta_k \langle k' \Lambda' | k \Lambda \rangle. \quad (\text{A6})$$

There is no ϕ_k dependence above, and since the twisted-photon states each have fixed Λ and θ_k , one can promote the states to twisted-photon states and obtain

$$\frac{\langle \kappa' m_\gamma k'_z \Lambda | J^3(\text{spin}) | \kappa m_\gamma k_z \Lambda \rangle}{\langle \kappa' m_\gamma k'_z \Lambda | \kappa m_\gamma k_z \Lambda \rangle} = \Lambda \cos \theta_k. \quad (\text{A7})$$

Continuing to the orbital angular momentum (OAM) piece, we need

$$\begin{aligned} & \langle \kappa' m_\gamma k'_z \Lambda | J^3(\text{OAM}) | \kappa m_\gamma k_z \Lambda \rangle \\ &= \langle \kappa' m_\gamma k'_z \Lambda | \int d^3x : \vec{A} \cdot \frac{\partial \vec{A}}{\partial \phi_\rho} : | \kappa m_\gamma k_z \Lambda \rangle. \end{aligned} \quad (\text{A8})$$

We pursue a different calculation here, still noting that within the normal ordering terms with two creation or two annihilation operators gives zero. For contributions where an

$a_{k\lambda}$ comes from $\partial \vec{A} / \partial \phi_\rho$ and an $a_{k'\lambda'}^\dagger$ comes from \vec{A} , the result is unchanged by inserting a vacuum intermediate state. The same is true for the reverse contribution. Hence

$$\begin{aligned} & \langle \kappa' m_\gamma k'_z \Lambda | J^3(\text{OAM}) | \kappa m_\gamma k_z \Lambda \rangle \\ &= 2 \int d^3x \langle \kappa' m_\gamma k'_z \Lambda | \vec{A} | 0 \rangle \langle 0 | \frac{\partial \vec{A}}{\partial \phi_\rho} | \kappa m_\gamma k_z \Lambda \rangle. \end{aligned} \quad (\text{A9})$$

We can use the results for the twisted-state wave functions, Eq. (11), and known Bessel function integrals to obtain

$$\begin{aligned} & \frac{\langle \kappa' m_\gamma k'_z \Lambda | J^3(\text{OAM}) | \kappa m_\gamma k_z \Lambda \rangle}{\langle \kappa' m_\gamma k'_z \Lambda | \kappa m_\gamma k_z \Lambda \rangle} \\ &= \frac{1}{2} m_\gamma \sin^2 \theta_k + (m_\gamma - \Lambda) \cos^4 \frac{\theta_k}{2} + (m_\gamma + \Lambda) \sin^4 \frac{\theta_k}{2} \\ &= m_\gamma - \Lambda \cos \theta_k. \end{aligned} \quad (\text{A10})$$

Combining the results,

$$\frac{\langle \kappa' m_\gamma k'_z \Lambda | J^3 | \kappa m_\gamma k_z \Lambda \rangle}{\langle \kappa' m_\gamma k'_z \Lambda | \kappa m_\gamma k_z \Lambda \rangle} = m_\gamma. \quad (\text{A11})$$

The total angular momentum projection in the direction of motion is precisely m_γ . The value of m_γ can be controlled in the laboratory by the means used to generate the beam of twisted photons, for example, by the use of spiral phase plates or computer-generated holograms [6].

-
- [1] R. Beth, *Phys. Rev.* **50**, 115 (1936).
[2] W. Heitler, *The Quantum Theory of Radiation* (Clarendon Press, Oxford, 1954).
[3] L. Allen, M. W. Beijersbergen, R. J. C. Spreeuw, and J. P. Woerdman, *Phys. Rev. A* **45**, 8185 (1992).
[4] G. Molina-Terriza, J. P. Torres, and L. Torner, *Nat. Phys.* **3**, 305 (2007).
[5] M. Uchida and A. Tomomura, *Nature* **464**, 737 (2010).
[6] A. Yao and M. Padgett, *Adv. Optics Photon.* **3**, 161 (2011).
[7] S. Sasaki and I. McNulty, *Phys. Rev. Lett.* **100**, 124801 (2008).
[8] A. Afanasev and A. Mikhailichenko, arXiv:1109.1603.
[9] E. Hemsing, A. Marinelli, and J. B. Rosenzweig, *Phys. Rev. Lett.* **106**, 164803 (2011).
[10] U. D. Jentschura and V. G. Serbo, *Phys. Rev. Lett.* **106**, 013001 (2011).
[11] U. Jentschura and V. Serbo, *Eur. Phys. J. C* **71**, 1571 (2011).
[12] A. Picón, J. Mompart, J. R. V. de Aldana, L. Plaja, G. F. Calvo, and L. Roso, *Opt. Express* **18**, 3660 (2010).
[13] A. Picón, A. Benseny, J. Mompart, J. R. V. de Aldana, L. Plaja, G. F. Calvo, and L. Roso, *New J. Phys.* **12**, 083053 (2010).
[14] B. S. Davis, L. Kaplan, and J. H. McGuire, *J. Opt.* **15**, 035403 (2013).
[15] R. Jáuregui, *Phys. Rev. A* **70**, 033415 (2004).
[16] T. Trueman and G. Wick, *Ann. Phys. (NY)* **26**, 322 (1964).
[17] L. Schiff, *Quantum Mechanics* (McGraw-Hill, New York, 1968).
[18] A. Davydov, *Quantum Mechanics* (Pergamon, New York, 1976).
[19] J. Bjorken and S. Drell, *Relativistic Quantum Mechanics* (McGraw-Hill, New York, 1964).