Circular dichroism of twisted photons in non-chiral atomic matter

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Abstract

We calculate the circular dichroism (CD) for the absorption of the twisted photons, or optical vortices, by atoms, caused by atomic excitation into discrete energy levels. The effects of photon spin on the rates and cross sections of atomic photo-excitation are considered. It is demonstrated that, although for electric dipole transitions the atomic excitation rates depend on the relative orientation of photon spin and orbital angular momentum (OAM), the resulting CD is zero. However, CD is nonzero for atomic transitions of higher multipolarity, and it is predicted to peak near the phase singularity in the optical vortex center. The effects remain large in a paraxial limit, where analytic expressions are provided. The predicted spin asymmetries are equivalent to OAM dichroism for the fixed photon spin.

Keywords: optical vortices, twisted photons, spin–orbit coupling, circular dichroism, photoexcitation, phase singularity

(Some figures may appear in colour only in the online journal)

1. Introduction and motivation

Circular dichroism (CD) is defined as differential absorption of left and right circularly polarized photons, and it is widely used in the analysis of materials that are non-symmetric under mirror transformation either through preferred chirality of the material structure or through magnetic phenomena [1].

Twisted photons, or optical vortices, carry orbital angular momentum (OAM) along their direction of propagation, and therefore they can be characterized by their own chirality (or topological charge); see e.g. [2] for a recent review. Spindependence of OAM photon flux enters through a spin–orbit interaction [3]. Interactions of the twisted photons with non-chiral nano-structures were reported in [4, 5], showing significant CD, at 80%–90% level, for varied topological charges. While the observed large CD can be understood in principle by realizing that left and right circular polarization states are not mirror-symmetric for twisted photons, the observed effect still awaits theoretical explanation, likely in terms of surface plasmon dynamics [5]. Theoretical predictions of CD were previously made for twisted x-rays in metals [6].

In the present work, we apply previously developed formalism for photo-excitation of atoms by the twisted photons [7, 8] and predict CD effects in the atomic matter. We take advantage of the observation [7, 9, 10] that the transition amplitudes of atomic photo-excitation with twisted photons can be presented in a simple factorized form in terms of plane-wave photon amplitudes, making our predictions applicable to a variety of atomic targets. We present the results as a function of the distance between a given atom (or ion) to the center of the optical vortex, that we define as an impact parameter b. The concept of an impact parameter is essential for describing the interaction of OAM light with atoms, and the corresponding formalism was extensively addressed in [11]. Precise measurements of ⁴⁰Ca⁺ ion excitations with the twisted light as a function of the impact parameter with sub-wavelength position resolution were performed recently [12] using an ion trap. For the most recent review of twisted-light interactions with atoms, see [13] and references therein.
Sections 2 and 3 of this paper review the formalism for twisted photons and for calculating atomic photoexcitation with the twisted photons, respectively. Section 4 introduces CD and discusses photon spin effects in the photoexcitation rates and cross sections, section 5 describes the evolution of the twisted-photon polarization caused by absorption, section 6 provides the analytic expressions in paraxial limit and section 7 offers some closing comments.

2. Definition of twisted-photon states

We define the twisted-photon states as non-paraxial Bessel beams according to [14, 15] that can be viewed as extensions of the nondiffractive Bessel modes described in [16, 17]. More detail is given in [7]. These states correspond to superposition of TE and TM Bessel modes introduced in [18]; see also the appendix of [8] for a detailed comparison.

A twisted photon state with symmetry axis passing through the origin, given as a superposition of plane waves and in Hilbert space, can be written as,

$$|\alpha m_z k \Lambda \rangle = \frac{\sqrt{\lambda}}{2\pi} \int \frac{dk}{2\pi} (-i)^m e^{im\phi_k} \left| k, \Lambda \right>.$$  \hspace{1cm} (1)

The component states on the right are plane-wave states, all with the same longitudinal momentum $k_z$, the same transverse momentum magnitude $\kappa = |k|$ and the same plane wave helicity $\Lambda$ (in the directions $k$). The angle $\phi_k$ is the azimuthal angle of vector $k$, and with the phasing shown, $m_z$ is the total angular momentum in the $z$-direction. We also define a pitch angle $\theta_k = \arctan(k_z/k_r)$, and $\omega = |k|$. The pitch angle $\theta_k$ was first introduced in the definition of Bessel beams in [16, 17], and it is related to Berry phase of the photon as [3]: $\phi_B = 2\pi(1 - \cos \theta_k)$. The phase singularity of this beam is located on the beam symmetry axis.

The electromagnetic potential of the twisted photon in coordinate space is

$$A_{\alpha m_z k \Lambda}^{\mu}(r, \tau) = \frac{\sqrt{\lambda}}{2\pi} e^{-i\omega t}$$

$$\times \int \frac{dk}{2\pi} (-i)^m e^{im\phi_k} \epsilon_{k,\Lambda}^{\mu} e^{i\mathbf{k} \cdot \mathbf{r}}.$$  \hspace{1cm} (2)

The polarization vectors are

$$\epsilon_{k,\Lambda}^{\mu} = e^{-i\Lambda \phi_k} \cos^2 \frac{\theta_k}{2} \eta_+^{\mu} + e^{i\Lambda \phi_k} \sin^2 \frac{\theta_k}{2} \eta_-^{\mu} + \frac{\Lambda}{\sqrt{2}} \sin \theta_k \eta_0^{\mu}$$  \hspace{1cm} (3)

with 4-dimensional unit vectors,

$$\eta_+^{\mu} = \frac{1}{\sqrt{2}} (0, \mp 1, -i, 0), \quad \eta_-^{\mu} = (0, 0, 0, 1).$$  \hspace{1cm} (4)

3. Plane-wave factorization for atomic photoexcitation with twisted photons

Here, we briefly review the formalism of atomic photoexcitation by the twisted photons worked out previously [7–10], leading to plane-wave factorization property of the twisted-photon absorption.

Consider the excitation by a twisted photon of an atom. The photon’s wave front travels in the $z$-direction and the axis of the twisted photon is displaced from the nucleus of the atomic target by some distance in the $x$–$y$ plane which we will call an impact parameter $b$, figure 1. The transition matrix
element is
\[ S_\rho = -i \int dt \langle n_f l_f m_f | H_\rho | n_i l_i m_i; \kappa \nu k, \Lambda \rangle, \]
where the non-relativistic interaction Hamiltonian is given by
\[ H_\rho = -\frac{e}{m_e} \tilde{\Lambda} \cdot \hat{p}, \]
and we use standard notation \((n, l, m)\) for the principal, orbital and magnetic quantum numbers of initial and final states of an atom.

It can be shown that [7, 9, 10] that for atomic excitation from the ground state \((l_i = m_i = 0)\) the above amplitude from equation (7) is proportional to the plane-wave amplitude \(\mathcal{M}^{\text{pw}}\) times Wigner \(d\)-functions that only depend on the pitch angle \(\theta_k\) and Bessel functions that define the amplitude dependence on the impact parameter \(b\). Here, the Bessel factor arises due to the azimuthal phase dependence of the twisted-photon and the excited atomic state, and Wigner \(d\)-functions is a result of a tilted quantization axis (by an angle \(\theta_k\)) with respect to the direction of beam propagation:
\[ |\mathcal{M}_{n_f l_f m_f}(b)| = \left| \frac{N}{2\pi} J_{m_f - m_i} (\kappa b) d_{n_f l_f}^{(1)} (\theta_k) \mathcal{M}^{\text{pw}}_{n_i l_i m_i}(\theta_k = 0) \right|. \]

The factorized form of the transition amplitude facilitates comparison of twisted-photon versus plane-wave absorption by atoms. It contains the details of atomic structure in a common-factor plane-wave amplitude \(\mathcal{M}^{\text{pw}}\), while the novel features arising from the phase and spatial structure of the twisted light are contained in Wigner and Bessel functions that enter independently of the specific details of atomic wave functions. When deriving the factorization property equation (9), we used the first-order Born matrix element that assumes the interaction proceeds in the linear regime, justifying representation of S-matrix as a linear superposition of plane-wave matrix elements. To further prove total angular momentum conservation under photo-absorption, we previously assumed that the atom is much smaller than the wavelength of absorbed light [8], but this assumption is not needed to derive equation (9). Therefore, as long as the linear interaction regime holds, the above formula is applicable to twisted-photon excitation of arbitrary quantum systems, such as atoms, molecules, ions, atomic nuclei, excitons or quantum dots. For example taking the limit \(\theta_k \rightarrow \pi/2\) in equation (9), we recover a similar factorization property recently derived for the absorption of polariton vortices, see equation (2) of [20].

4. Spin-dependence and CD of twisted-photon absorption

The twisted-photon flux depends on the sign of \(\Lambda\) defining the handedness of plane-wave photons that form a given Bessel beam, and this dependence was discussed previously by Bliokh and collaborators in the context of spin–orbit interaction of light [3, 21].
presented as rates (or Rabi frequencies as in [12]) normalized to the total laser-beam power within an aperture that is much wider than the wavelength of light. In this case, a relevant observable would be \( \Lambda \)-dependence of the photo-excitation rate that was analyzed theoretically in [7] (for the case of leading \( E_1 \) transitions).

We define photon-spin asymmetry of the photo-excitation rate similarly to CD,

\[
A_{\Lambda}(\bar{m}, \ell_f) = \frac{\Gamma^{(\mathbb{S},+)}_{n_f/l_f; \Lambda=1} - \Gamma^{(\mathbb{S},-)}_{n_f/l_f; \Lambda=-1}}{\Gamma^{(\mathbb{S},+)}_{n_f/l_f; \Lambda=1} + \Gamma^{(\mathbb{S},-)}_{n_f/l_f; \Lambda=-1}}.
\]

The results are presented in figure 4 for the atomic transitions into the states of \( \ell_f = 1, 2 \) and 3 caused by the photons with topological charges \( \bar{m}_s \) to 3. We can see that the spin asymmetry of rates behaves differently from CD: the asymmetry is negative within the distance of about one wavelength near the optical vortex center and, with a few exceptions, reaches a value of \(-1\) at the center. It means that the transitions at the vortex center are mainly caused by the twisted photons whose spin and OAM projections (\( \Lambda \) and \( m_r \)) have opposite signs, i.e. they are are anti-aligned. An apparent difference from positive-sign CD is due to the fact that there is a relatively denser flux of the anti-aligned twisted photons at the vortex center for the same overall beam power.

If the atom’s position is not resolved, we would have to integrate over the position, resulting into zero spin asymmetry \( A_{\Lambda} \), similarly to the above result for CD. It implies that observation of spin-asymmetric absorption of twisted light by atomic matter requires localization of the target atoms within about light’s wavelength. It can be achieved, for example, by using nano-sized apertures, well-localized ions in Paul traps, or mesoscopic targets.

5. Evolution of the twisted-light polarization under propagation in matter

Above predictions of nonzero CD for the twisted light being absorbed by atoms would lead to the evolution of twisted-photon polarization states. Indeed, let us represent an arbitrary polarization state as a superposition of \( \Lambda = 1 \) and \( \Lambda = -1 \) spin states, or left- and right-circularly polarized states, for a
given topological charge:

$$c_{\pm} = c_{\pm}|n\mathbf{m}, k; \Lambda| = c_{\pm}|n\mathbf{m}, k; \Lambda| = 1,$$

where $c_{\pm}$ are complex coefficients. Their dependence on the propagation distance $z$ is controlled by the attenuation coefficients $\mu_{\pm}$, that in turn are proportional to the photoabsorption cross sections $\sigma_{\pm}$, with $n$ a number of atoms per unit volume, with the expressions for $\sigma_{\pm}$ coming from equation (10). Since we are interested in comparison with the plane-wave propagation, we can express the attenuation coefficients in terms of their plane-wave values and the cross section ratios $r_{\pm}^b$ introduced in [7, 10]:

$$\mu_{\pm} = \mu_{\pm}^p \frac{\sigma}{\sigma_{\pm}} = \mu_{\pm}^p r_{\pm}^p (b),$$

where the plane-wave attenuation coefficient $\mu_{\pm}^p$ is independent of $\Lambda$ for the isotropic atomic matter, while the factor $r_{\pm}^p (b)$ depends on $\Lambda$ and on the impact parameter $b$. Then

$$c_{\pm}(z) = c_{\pm}(0) e^{-bz/2} = c_{\pm}(0) e^{-\mu_{\pm}^p r_{\pm}^p (b)/2}.$$

For the case of electric dipole transitions $l_f = 1$, it follows from equations (6) and (9) that $r_{\pm}^p (b) = 1/\cos \theta_f$, independently of $\Lambda$ and $b$ [7, 9, 10]. Therefore, the coefficients $c_{\pm}(z)$ have the same $z$-dependence resulting in no evolution of the twisted-photon polarization due to atomic absorption via electric-dipole transitions. However for higher-multipole transitions into the states $l_f > 1$ the factors $r_{\pm}^p (b)$ depend on the photon spin projection due to CD as defined in equation (12) (since the plane-wave cross section is a constant and it cancels in the ratio).

For example, let us consider a superposition of $\Lambda = 1$ and $\Lambda = -1$ spin states for the same topological charge $\mathbf{m}_z = 1$, and assume the coefficients to be real and initially equal: $c_{+}[z = 0] = c_{-}[z = 0]$, where the field potential with a given $\Lambda$ is defined by equation (5). It results in a state with almost 100% linear polarization of transverse fields in the central region of the vortex (except for the small region near the node of Bessel function $J_0$). However, even for small values of propagation distance $z$, the optical vortex develops 100% circular polarization in the vortex center (C-point), and this region broadens as the beam passes further through the atomic matter. This prediction is shown in figure 5, where we used standard definitions for Stokes parameters $S_{0-3}$ [22]. Development of C-type polarization singularity at the vortex center as a result of beam propagation can be observed in a dedicated experiment.

It should be noted that here we only considered the effects from photon absorption that result in CD. An additional effect would be forward scattering of the twisted photons, see e.g. [23]. Corresponding spin dependence would result in circular birefringence showing in rotation of polarization plane of the linearly-polarized twisted light due to spin dependence of refractive index.

### 6. Spin-dependence and CD in paraxial limit

Numerical calculations of spin-dependent observables for the twisted light with a fixed value of $\mathbf{m}$, reveal smooth transition to the paraxial limit $\theta_f \to 0$, in which the quantity $\mathbf{m}$, unambiguously determines OAM projection (or topological charge) of the twisted photon. Actually, as seen in figure 3, there is little dependence on this angle for $\theta_f \leq 0.25$ rad. This small-angle behavior prompts us to apply small-angle Taylor expansion for the expressions of CD and $\Lambda_\Lambda$ using explicit formulae equations (6), (9). Noticing that the argument of Bessel functions is $\kappa b = k \sin \theta_f b \approx k b \theta_f$ and defining $kb \equiv x$, we consider $\Lambda$-dependence of the photon flux.
where in general all three terms of equation (6) appear to be of the same order in $\theta_k$ for the anti-aligned spin and OAM.

This expression closely matches rate asymmetries in figure 4(a) for moderately small pitch angles $\theta_k \leq 0.25$. It is remarkable that while the beam waist size strongly depends on the angle $\theta_k$ (which in turn relates to Berry phase [3]), the spin asymmetry in the paraxial limit depends only on the topological charge $m$. 

We can use the same approach to determine the spin asymmetries of photoexcitation rates for higher transition multipolarities, that in addition requires small-angle expansion of Wigner $d$-functions in equation (9). For example, for the quadrupole transitions caused by $m_z = 1$ beam, we identify the terms that are leading-order in small $\theta_k$-expansion:

$$m_z = 2, \Lambda = 1,$$
$$\mathcal{M}(m_f = 2) \propto J_0(xb) \cdot d_{21}^{(2)}(\theta_k) \approx \theta_k,$$
$$\mathcal{M}(m_f = 1) \propto J_1(xb) \cdot d_{21}^{(2)}(\theta_k) \approx k\theta_k/2,$$

Considering the amplitudes and summing over all magnetic quantum numbers $m_f$ according to equations (10) and (14), we obtain:

$$\lim_{\theta_k \to 0} A_{\Lambda}^{(m_z=1,m_f=2)} = \frac{-1}{5 + x^2},$$

that results in $-20\%$ asymmetry in the vortex center, in agreement with exact results from figure 4(b). We can trace the factors yielding this value of the asymmetry to the differences between Wigner $d$-functions $d_{21}^{(2)}(\theta_k)$ and $d_{11}^{(2)}(\theta_k)$ multiplying non-vanishing transition amplitudes in the vortex center: the latter is larger by a factor $\sqrt{3}/2$ in a small-angle limit. Experimental observation of this difference in the excitation amplitudes can be made by analyzing normalized Rabi frequencies in ion traps, in a setup similar to [12].

Taking the expressions for CD equation (12), we obtain in the paraxial limit, for example:

$$\text{CD}^{(m_z=-1,m_f=-2)} = -\frac{4}{x^4 + 6x^2 + 4}.$$  

Other analytic expressions in the paraxial limit for different values of the topological charge $m_z$ and the excited-atom OAM $L$ are listed in the appendix.

7. Summary and discussion

In this work, we applied previously developed theoretical formalism [7–9] to analyze photoexcitation of an atom by Bessel beams with OAM and with different orientations of
photon spin. We found that both the photoexcitation rates and cross sections of twisted-photon absorption show strong dependence on the relative orientation of spin and OAM along the beam propagation direction. They can be observed by fixing the spatial structure of the beam (i.e., OAM) and flipping circular polarization with quarter-wave plates. From the parity considerations, it would be equivalent, up to an overall sign, to fixing the circular polarization and analyzing dependence on the sign of OAM projection on the beam propagation direction. Therefore, our calculations predict both circular and OAM dichroism of the twisted light. Due to the factorization property of the twisted-light photoexcitation amplitudes in the first Born approximation equation (9), the plane-wave matrix elements cancel in the spin asymmetries (due to parity conservation), yielding the results independent of the internal structure of the atomic target.

Since the rates of electric dipole transitions are proportional to the local energy flux, the corresponding CD is zero. However, position-dependent photoexcitation rates show strong dependence on the photon spin, in manifestation of spin–orbit interaction of light, see [3, 21]. The corresponding rate asymmetry becomes independent of the pitch angle \( \theta_k \) for moderately small angles below about 0.25 rad. This observation provides a possible method for determination of the topological charge of optical vortex. For the higher-multipole transitions with excitation of \( l_f > 1 \) atomic states, the spin asymmetries are large near the optical vortex center; the asymmetries die off at distances about one wavelength from the vortex center. For electric quadrupole transitions caused by the beams of topological charge \( m_\ell = 1 \), the maximum spin asymmetry of the excitation rates is predicted to be \(-20\%\), that may be observed in the experimental setup similar to [12] by analyzing Rabi frequencies for individual electronic transitions in an ion trap.

We demonstrated that the OAM light, through higher-multipole transition, shows CD even in isotropic atomic matter. In absence of OAM of light—for the light beams of zero topological charge—such property would signal parity non-conservation, a phenomenon revealed in atoms at about \( 10^{-7} \)–\( 10^{-8} \) level due to neutral weak currents [24]. However, as we have shown in this paper, CD can reach 100\% in atomic photoexcitation by OAM light, uniquely indicating presence of beam’s OAM, and showing the impact parameter dependence that is distinctly different for different OAM values. While our predictions strongly depend on the beam’s topological charge, they are practically independent of other beam parameters (such as Berry phase) that are important for non-paraxial beams. These observations can provide a diagnostic tool for accessing beam’s topological charge by observing excitations of single atoms. Such a method can be realized by extending an approach of [12], whereby the strength of various Zeeman-split transition can be measured. In realizing OAM quantum memory [13, 25], the storage and recall of photon states with opposite helicities is important; our analysis can therefore help with implementation of quantum computing with OAM light.

We believe the present analysis of spin dependence of light absorption in atomic matter will be instrumental for quantum computing applications, optical communications, imaging, and characterization of twisted light in a broad range of wavelengths.

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**Appendix. Expressions for spin asymmetries in a paraxial limit**

Here we present the expressions for CD and \( A_\lambda \) for small values of the pitch angle \( \theta_k \to 0 \), with \( x \equiv k \cdot b \).

**Electric quadrupole transitions \( l_f = 2 \)

*Rate Asymmetries*

\[
A_{\lambda}^{(m_\ell = 1, l_f = 2)} = -\frac{1}{x^2 + 5},
\]

\[
A_{\lambda}^{(m_\ell = 2, l_f = 2)} = -\frac{2(2x^2 + 9)}{x^4 + 20x^2 + 18},
\]

\[
A_{\lambda}^{(m_\ell = 3, l_f = 2)} = -\frac{9(x^4 + 18x^2 + 8)}{x^6 + 45x^4 + 162x^2 + 72},
\]

\[
A_{\lambda}^{(m_\ell = 4, l_f = 2)} = -\frac{8(2x^4 + 81x^2 + 144)}{x^8 + 80x^6 + 648x^4 + 1152}. \tag{25}
\]

**Circular Dichroism**

\[
CD^{(m_\ell = 1, l_f = 2)} = \frac{4}{x^4 + 6x^2 + 4},
\]

\[
CD^{(m_\ell = 2, l_f = 2)} = \frac{48x^2 + 32}{x^6 + 24x^4 + 84x^2 + 32},
\]

\[
CD^{(m_\ell = 3, l_f = 2)} = \frac{36(5x^2 + 16)}{x^6 + 54x^4 + 504x^2 + 720},
\]

\[
CD^{(m_\ell = 4, l_f = 2)} = \frac{64(7x^2 + 54)}{x^6 + 96x^4 + 1744x^2 + 5760}. \tag{26}
\]

**Electric octupole transitions \( l_f = 3 \)

*Rate Asymmetries*

\[
A_{\lambda}^{(m_\ell = 1, l_f = 3)} = -\frac{1}{x^2 + 11},
\]

\[
A_{\lambda}^{(m_\ell = 2, l_f = 3)} = -\frac{4x^2 + 42}{x^4 + 44x^2 + 102},
\]

\[
A_{\lambda}^{(m_\ell = 3, l_f = 3)} = -\frac{9(x^4 + 42x^2 + 80)}{x^6 + 99x^4 + 918x^2 + 720},
\]

\[
A_{\lambda}^{(m_\ell = 4, l_f = 3)} = \frac{8(2x^6 + 189x^4 + 1440x^2 + 540)}{x^8 + 176x^6 + 3672x^4 + 11520x^2 + 4320}. \tag{27}
\]
Circular Dichroism

\[
\text{CD}(\sigma_+, l=3) = \frac{10}{x^4 + 12x^2 + 10},
\]
\[
\text{CD}(\sigma_-, l=3) = \frac{40(3x^4 + 8x^2 + 3)}{x^8 + 48x^6 + 264x^4 + 320x^2 + 120},
\]
\[
\text{CD}(\pi, l=3) = \frac{90(5x^4 + 64x^2 + 108)}{x^8 + 108x^6 + 1746x^4 + 7200x^2 + 9720},
\]
\[
\text{CD}(\sigma_-, l=3) = \frac{160(7x^4 + 216x^2 + 945)}{x^8 + 192x^6 + 6304x^4 + 57600x^2 + 159840}.
\]

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