

**OPTICAL PHYSICS** 

# Excitation of E1-forbidden atomic transitions with electric, magnetic, or mixed multipolarity in light fields carrying orbital and spin angular momentum [Invited]

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Photons carrying a well-defined orbital angular momentum have been proven to modify spectroscopic selection rules in atomic matter. Excitation profiles of electric quadrupole transitions have been measured with single trapped  ${}^{40}Ca^+$  ions for varying polarizations. We further develop the photo-absorption formalism to study the case of arbitrary alignment of the beam's optical axis with respect to the ion's quantization axis and mixed multipolarity. Thus, predictions for *M*1-dominated  ${}^{40}Ar^{13+}$ , E3-driven  ${}^{171}Yb^+$  and  ${}^{172}Yb^+$ , and B-like  ${}^{20}Ne^{5+}$  are presented. The latter case displays novel effects, coming from the presence of a strong photon-magnetic dipole coupling. © 2019 Optical Society of America

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# **1. INTRODUCTION**

The interest of scientists in vortex-like dislocations in various media goes back to 1970s [1,2]. For historic and topical reviews, we would like to refer the reader to such sources as [3,4]. The ability of twisted photon beams to transfer orbital angular momentum (OAM) as an extra degree of freedom, first shown in Ref. [5], has largely influenced our understanding of light, e.g., [6–13]. It made OAM-carrying laser beams a popular research subject in optical and quantum communications and information security [14–18]. Nontrivial topological structure of such OAM-carrying optical vortices, paired with progress in photonics and nanotechnology [19], resulted in proposals and subsequent implementations of novel devices and techniques [20–25].

Over years, significant progress has been made towards understanding the mechanisms of twisted light interaction with matter. It became clear that in order to detect transfer of photon OAM to the internal degrees of freedom of electron configurations, one needs to consider transitions driven by contributions higher than the electric dipole E1. This is the nontrivial electromagnetic field distribution in the beam vortex, which bound electrons with high angular momentum are sensitive to [26,27]. It has been shown for molecular [28] and atomic [29] systems, as well as semiconductor heterostructures [30,31].

In this paper, we focus our attention on OAM photons interacting with single trapped ions. The possibility to enhance and control weak atomic transitions was discussed in Ref. [32], where the authors had foreseen OAM modes being used in high-precision atomic spectroscopy. Particular interest in the field has been paid to light–matter interactions in the case of beams carrying a well-defined OAM [33–36]. In the past, a detailed experimental study demonstrated the transfer of photon OAM to the valence electron of a single trapped <sup>40</sup>Ca<sup>+</sup> ion [11,37]. This progress was based on the technologies developed for ion-trap quantum computing [38].

Now we propose extending these methods to include atomic transitions in ions with complex level structure such as Yb<sup>+</sup>. We also study selection rules in highly charged ions (HCIs) as  $Ar^{13+}$  and  $Ne^{4+}$ . We aim to stimulate experimental studies on these transitions with OAM light, and have chosen cases that have already been experimentally studied using Gaussian beams [39–41] or have accessible optical transitions. The theory considers arbitrary alignment of the field vector with respect to the atomic quantization axis and takes hyperfine splitting into account, extending previous investigations [36].

The paper is structured as follows: the theoretical approach is described in Section 2. It is built on the formalism for Bessel (BB) and Bessel-Gauss (BG) presented in Refs. [32,36,37], where multipole expansion of the plane wave contribution has been used, similar to [10]. In Section 3, we compare our findings with data obtained in <sup>40</sup>Ca<sup>+</sup> ions as a test case for our theory. In this case, the next-to-the-leading M3 multipole is strongly suppressed, compared to the dominating E2. Previously, the authors in Ref. [11] used semiclassical formalism, while we follow a purely quantum approach of evaluating the interaction strength in terms of photo-absorption transition amplitudes. While certain data we use here were already published before, the data for various photon polarization states are presented to the public for the first time. Section 4 is devoted to theoretical predictions for the ions with strong electric, magnetic, and mixed multipolarities. We study the M1-dominant  $P_{1/2} \rightarrow P_{3/2}$  transition in <sup>40</sup>Ar<sup>13+</sup>, the E3-driven transition in 171Yb or 172Yb [42–45], and the mixed M1 + E2 multipolarity transition in <sup>20</sup>Ne<sup>5+</sup> [46]. The summary is provided in Section 5.

# 2. FORMALISM

For twisted photons in a BB whose vortex line is displaced from the center of the target atom by a distance b, and where the atomic states are quantized along an axis parallel to the beam propagation direction, the transition amplitude M reads [29,36]

$$M_{m_{f}m_{i}\Lambda}^{(\text{BB})}(b;\theta_{z}=0) = Ai^{m_{f}-m_{i}-2m_{f}}e^{i(m_{f}+m_{i}-m_{f})\phi_{b}}J_{m_{f}-m_{f}+m_{i}}(\kappa b)$$

$$\times \sum_{m'_{f}m'_{i}}d_{m_{f},m'_{f}}^{j_{f}}(\theta_{k})d_{m_{i},m'_{i}}^{j_{i}}(\theta_{k})M_{m'_{f}m'_{i}\Lambda}^{(\text{pw})}(0).$$
(1)

This amplitude is proportional to experimentally measured transition strength  $\Omega_R = \Omega_{\text{Rabi}}$ , e.g., [11]. A denotes an overall normalization factor. The initial and final atomic states have quantum numbers  $\{j_i, m_i\}$  and  $\{j_f, m_f\}$  (where  $j_{i,f}$  are total angular momenta of the electronic states, with orbital angular momenta  $l_{i,f}$ ), and  $m_{\gamma}$  is the projection of the photon beam's total angular momentum on the direction of the beam propagation, the z axis.  $J_m(\kappa b)$  is a Bessel function with the argument given by the impact parameter  $\boldsymbol{b} = \{b \cos \phi_b, b \sin \phi_b, 0\},\$ and the transverse momentum magnitude  $\kappa = \sqrt{k^2 - k_z^2}$ . The  $d_{m,m'}^{j}(\theta_k)$  are Wigner rotation matrices evaluated at the pitch angle  $\theta_k$  [47].  $M^{(pw)}$  is the transition amplitude for a plane wave traveling in the z direction with helicity  $\Lambda = \pm 1$ . We will also define  $\ell_{\gamma}$  from the identity  $m_{\gamma} \equiv \ell_{\gamma} + \Lambda$ , which can be interpreted as the photon's OAM in paraxial approximation.

In our practical considerations, we have used a BG mode, which is a BB with peripheral behavior suppressed by a Gaussian factor to better fit experimental conditions. Additionally, when the static magnetic field that defines the quantization axis of the atomic states points in a general direction  $\{0, \theta_z, \phi_z\}$ , one has

$$\mathcal{M}_{m_{f}m_{i}\Lambda}^{(\mathrm{BG})}(b;\theta_{z}) = e^{-b^{2}/w_{0}^{2}} e^{-i(m_{f}-m_{i})\phi_{z}} \\ \times \sum_{m_{f}'m_{i}'} d_{m_{f}m_{f}'}^{j}(\theta_{z}) d_{m_{i}m_{i}'}^{j_{i}}(\theta_{z}) \mathcal{M}_{m_{f}',m_{i}'\Lambda}^{(\mathrm{BB})}(b;\theta_{z}=0),$$
(2)

where  $w_0$  is the Gaussian width of the beam (beam waist), as defined in, e.g., [48].

A plane wave photon can be expanded in terms of spherical waves. Each spherical wave has total angular momentum j, and the plane wave amplitude can be expressed in terms of spherical multipoles [32],

$$M_{m_f m_i \Lambda}^{(\text{pw})}(0) = -\sum_{j=1}^{\infty} i^{j+\mu} \sqrt{\frac{4\pi (2j+1)}{2j_f + 1}} \Lambda^{\mu+1} C_{j_i m_j \Lambda}^{j_f m_f} M_{j\mu}.$$
 (3)

The  $C_{j_j m_i j\Lambda}^{j_f m_f}$  are Clebsch–Gordan coefficients, and  $M_{j\mu}$  is the spherical amplitude of order *j* and multipolarity  $\mu$ . Magnetic multipoles are described with  $\mu = 0$  and electrical ones with  $\mu = 1$ . We conveniently simplify by writing  $M_{j0} = Mj$  and  $M_{j1} = Ej$ .

While the sum in Eq. (3) is formally infinite, typically only two amplitudes contribute for given atomic states, and often one of them is small enough to neglect. When an atom has nuclear spin I, magnetic hyperfine interaction may significantly contribute to the atom photo-excitation. In case of a nondeformed nucleus, such as <sup>171</sup>Yb<sup>+</sup>, the nuclear electric quadrupole does not contribute, and the photon angular momentum couples to the total atomic angular momentum F = J + I(TAM). Hence, in Eq. (3), one requires the substitution of the principal quantum numbers  $|j_f m_f\rangle \rightarrow |j_f I F_f m_f^{(f)}\rangle$ and  $|j_i m_i\rangle \rightarrow |j_i I F_i m_i^{(f)}\rangle$ , e.g., [49], and of the expansion coefficients,

$$\sqrt{\frac{1}{2j_f+1}} C_{j_i m_j \Lambda}^{j_f m_f} \rightarrow (-1)^{j_f+I+F_i-j} \sqrt{(2F_i+1)} C_{F_i m_i^{(f)} j \Lambda}^{F_f m_f^{(f)}} \times \begin{cases} j_f & F_f & I \\ F_i & j_i & j \end{cases},$$
(4)

where 6*j* are Wigner coefficients, as defined in Ref. [50].

#### 3. <sup>40</sup>Ca<sup>+</sup>: THEORY AND EXPERIMENT

We consider the atomic transition  $4^2S_{1/2} \rightarrow 3^2D_{5/2}$  in a single trapped  ${}^{40}\text{Ca}^+$  ion. The coupling scheme, outlined in Eqs. (3) and (4), allows two multipole contributions: *M3* and *E2*. However, *M3* is much weaker than *E2*. This is due to *M3* contributing in the second-to-the-leading order in the Taylor expansion of  $e^{i\mathbf{k}\cdot\mathbf{r}}$ , while *E2* arises in first, along with *M1*, e.g., [51]. To the best of the authors' knowledge, there are also no spectroscopic effects related to the presence of *M3*. So the plane wave amplitude  $M^{(\text{pw})}$  can be written as

$$M_{m_f m_i \Lambda}^{(\text{pw})} \simeq C_{1/2 m_i 2 \Lambda}^{5/2 m_f} E2.$$
 (5)

The interaction strength for a beam of definite helicity  $\Lambda$  and TAM  $m_{\gamma}$  can then be calculated using Eq. (2). One obtains

$$M_{m_{f}m_{i}\Lambda}^{(BG)}(b;\theta_{z}=0)$$

$$\simeq Ai^{m_{f}-m_{i}-2m_{\gamma}}e^{-b^{2}/w_{0}^{2}}e^{i(m_{\gamma}+m_{i}-m_{f})\phi_{b}}J_{m_{\gamma}-m_{f}+m_{i}}(\kappa b)$$

$$\times \sum_{m_{i}^{'}}d_{m_{f},m_{i}^{'}+\Lambda}^{j_{f}}(\theta_{k})d_{m_{i},m_{i}^{'}}^{j_{i}}(\theta_{k})C_{j_{i}m_{i}^{'}2\Lambda}^{j_{f}m_{i}^{'}+\Lambda}E2.$$
(6)

We now compare these results with interaction strengths measured with a single trapped  ${}^{40}Ca^+$  ion, Zeeman-split in a static magnetic field [11]. In the experiment, an ion was trapped in a microstructured segmented Paul trap with the 5 nm position resolution. The interaction strength was measured as a Rabi frequency as a function of the position of the ion with respect to the beam (the impact parameter *b*). For the details about the experimental setup and trapping techniques, we refer the interested reader to, e.g., [11].

First we consider data taken with the angle between the quantization axis and beam propagation vector  $\theta_z = \pi/4$ . We consider two datasets. One, shown in Fig. 1, where excitation profiles are measured as a function of the varying polarization. And a second one, shown in Fig. 2(h) and Figs. 3(d) and 3(e), where detailed profiles are measured for left-circular, horizontal (H), and vertical (V) polarizations of light, defined with respect to the incidence plane formed by the direction of the static magnetic field and the beam axis.

For both datasets in Figs. 1 and 3, we consider the transitions  ${}^{2}S_{1/2} \rightarrow {}^{2}D_{5/2}$  between the Zeeman-split levels:  $m_{i} = 1/2$  into  $m_{f} = 3/2$ . The polarization angles for the measurements were extracted by comparing the optical response in experiments and those generated in the simulation. We describe the polarization vector of the beam as

$$\hat{e} = e^{i\delta}(\hat{e}_{-}\cos(\alpha/2) - \hat{e}_{+}\sin(\alpha/2)e^{-i2\delta}),$$
 (7)

where the polar angle  $\alpha$  and phase retardation  $\delta$  range in the interval  $[0, \pi]$ . The convention used here for right-circular  $\hat{e}_{-}$  and left-circular  $\hat{e}_{+}$  polarization states is  $\hat{e}_{\pm} = \frac{1}{\sqrt{2}} \{ \mp 1, -i, 0 \}$ . The H state is obtained with  $\alpha = 90^{\circ}$  and  $\delta = 0$ , and the V state is obtained using  $\alpha = 90^{\circ}$  and  $\delta = 90^{\circ}$ .

For the case of linearly polarized incoming photons, this can be rewritten as

$$M_{m_f m_i H/V}^{(BG)}(b;\theta_z) = \cos(\pi/4) M_{m_f,m_i-1}^{(BG)}(b;\theta_z) - \sin(\pi/4) e^{-i2\delta} M_{m_f,m_i-1}^{(BG)}(b;\theta_z).$$
 (8)

Then we substitute the Eq. (6), rotated by angle  $\theta_z$ , as in Eq. (2). The nonzero phase difference, coming from exponential factor  $\exp[i(m_{\gamma} + m_i - m_f)\phi_b]$ , introduces a new parameter  $\phi_b$ . In our simulations, we are tuning this polar angle  $\phi_b$ , responsible for the azimuthal orientation of the ion with respect to the plane formed by the beam propagation axis and constant magnetic field.

The calculation of the interaction strength involves two rotations performed sequentially: (1) the rotation of the plane wave photon by the pitch angle  $\theta_k$  and (2) the rotation of the twisted beam over the angle  $\theta_z$ . While the  $\phi_b$ -dependent phase drops out on the level of Rabi frequencies  $\Omega_R \propto$  $|M_{m_f m_i \Lambda}|$  for definite helicity states, the same phase factor  $\exp(im_\gamma \phi_b)$  becomes observable for H and V states. To control the polarization of the beam, we used a set of wave plates [11]. By rotating a half-waveplate, the polarization state is varied along a meridian in the Poincaré sphere.

In Fig. 2(h) and Figs. 3(d) and 3(e), we show a comparison of the theoretical prediction with the experimental results for beams with different OAM  $\ell_{\gamma} = 0, 1, 2$  with either vertical or horizontal polarization. Some features of the data can be understood by analytic expansions of Eq. (1) for the various cases.



**Fig. 1.** Contour plots of the normalized transition strength as a function of polarization and impact parameter for  $4^2S_{1/2} \rightarrow 3^2D_{5/2}$  in a single  ${}^{40}\text{Ca}^+$ . From left to right, the first and second subplots are for  $\ell_{\gamma} = 0$  theory and experimental data, respectively. The third and fourth are for  $\ell_{\gamma} = 1$  theory and experimental data, respectively. The theory prediction is fitted using  $\theta_k = [0.075, 0.095]$  rad and  $\phi_b = [-0.62, -0.3]$  rad for  $\ell_{\gamma} = 0$  and 1, respectively. Red lines indicate pure vertical polarization.

For small impact parameter  $b \rightarrow 0$ , the Bessel function will collapse into delta function, and the projection of the photon's total angular momentum  $m_{\gamma}$  will be transferred into the internal degrees of freedom of the target atom,  $\Delta m = m_f - m_i = m_{\gamma}$  [36,37,52]. In such a case, the interaction strengths only depend on the pitch angle  $\theta_k$  and the alignment angle  $\theta_z$ ,

$$M_{3/2 \ 1/2 \ H}^{(\mathrm{BG})(\ell_{\gamma}=0)} \propto i(5\theta_k^2 - 4)\cos(2\theta_z)$$
  
$$M_{3/2 \ 1/2 \ H}^{(\mathrm{BG})(\ell_{\gamma}=1)} \propto 2\theta_k(1 + 4 \cos \theta_z)\sin \theta_z$$
  
$$M_{3/2 \ 1/2 \ H}^{(\mathrm{BG})(\ell_{\gamma}=2)} \propto i\frac{3}{\sqrt{2}}\theta_k^2(\cos \theta_z + \cos 2\theta_z)$$

the beam, the only nonvanishing transitions are those in which the photon OAM matches the change in magnetic number  $\Delta m = m_{\gamma}$ . However, it is interesting to note that the  $\Delta m =$ 3 transition is null even if the conservation conditions are met. This is due to an extra selection rule in which  $|\Delta m| \leq$  $|\Delta l| = 2$  for *E*2 transitions.

$$M_{3/2 \ 1/2 \ V}^{(\text{BG})(\ell_{\gamma}=0)} \propto i(5\theta_{k}^{2}-4)\cos(\theta_{z}),$$

$$M_{3/2 \ 1/2 \ V}^{(\text{BG})(\ell_{\gamma}=1)} \propto 2\theta_{k}(2\cos\theta_{z}-1)\sin\theta_{z},$$

$$M_{3/2 \ 1/2 \ V}^{(\text{BG})(\ell_{\gamma}=2)} = -M_{m(m,H}^{(\text{tw})}(\ell_{\gamma}=2).$$
(9)

From the equations above, one can see that the horizontal polarization is completely suppressed in the vortex center when  $\theta_z = \pi/4$  and  $\ell_\gamma = 0$ . In 45 deg alignment, this is a signature of transitions with dominant *E*2. This is due to their sensitivity to the field gradients [11] and generation of geometry-dependent terms due to rotation of the quantization axis [Eq. (2)]. The dependence on the alignment angle was previously discussed in Refs. [53–55].

When performing the expansion for different OAM expressions in Eq. (9), there is the crucial observation to make: the plane wave transition amplitudes of different helicity do not contribute on equal basis as the OAM goes up. While for  $\ell_{\gamma} = 0, 1$ , left-circular ( $\Lambda = 1$ ) and right circular ( $\Lambda = -1$ ) contribute symmetrically, for  $\ell_{\gamma} = 2$ , only right-circular components contribute into the absorption matrix element at  $b \rightarrow 0$ . This leads to the effect of local circular dichroism in isotropic targets. This phenomenon is position dependent and gets stronger for beams with higher vorticity, until all the contributions become extinct at the vortex center. This result confirms and reinforces our earlier findings [56].

In Fig. 1, we show the excitation profiles for a *varying* polarization for a Gaussian beam and one with  $\ell_{\gamma} = 1$ . The polarization state is scanned along a meridian in the Poincaré sphere parameterized by angle  $\alpha$ , as defined in Eq. (7). We see that the theoretical model reproduces all the main features, including the breaking of radial symmetry for polarizations that are not purely linear. The fit parameters are the beam waist  $w_0$ , the pitch angle  $\theta_k$ , and the azimuthal angle  $\phi_b$ . The fit was performed using the whole dataset. An overall normalization constant and pitch angle were picked for each profile independently, while the waist  $w_0 = 9 \ \mu m/\lambda$  and azimuthal angle were constrained to be the same throughout. We obtain  $\theta_z = 45^\circ \pm 5^\circ$  and  $\delta = 0 \pm 0.02$ . The polarization patterns are strongly dependent on angle  $\phi_b$ . In particular, when  $\phi_b \rightarrow -\phi_b$ , one would get a picture, mirror-symmetric to Fig. 1.

Finally in Figs. 2(b), 2(e), and 2(h), we show excitation profiles for alignment angle  $\theta_z = 0$  and left-circular polarization  $\Lambda = 1$ . We plot the results for different OAM  $\ell_{\gamma} = 0, 1, 2$ and different changes in magnetic number  $\Delta m = 1, 2, 3$ . At this angle, the conservation of the projection of angular momentum for  $b \rightarrow 0$  is observed. This is why, at the center of

# 4. PREDICTIONS FOR *M*1, *E*3, AND MIXED MULTIPOLARITY

In this section, we present theoretical predictions of structured light field excitation for atomic ions featuring electric and magnetic multipole transitions of interest.

# A. <sup>40</sup>Ar<sup>13+</sup>M1-Driven Transition

In the highly charged ion  ${}^{40}\text{Ar}{}^{13+}$ , the transition  $P_{1/2} \rightarrow P_{3/2}$  is an example where the *M*1 amplitude dominates while the *E*2 is suppressed by 6 orders of magnitude [57]. The relevant plane wave amplitudes from Eq. (3) are then

$$M^{(\text{PW})}_{(3\Lambda/2)(\Lambda/2)\Lambda} = \Lambda\sqrt{3\pi}M1,$$
  

$$M^{(\text{BB})}_{(\Lambda/2)(-\Lambda/2)\Lambda} = -\Lambda\sqrt{\pi}M1.$$
(10)

Following the procedure from the previous sections and under the condition of small b, one gets the transition strengths,

$$M_{3/2 \ 1/2 \ H}^{(\text{BG})(\ell_{\gamma}=0)} \propto -i \left(1 - \frac{\theta_{k}^{2}}{4}\right) \qquad M_{3/2 \ 1/2 \ V}^{(\text{BG})(\ell_{\gamma}=0)} \propto i \left(1 - \frac{\theta_{k}^{2}}{4}\right) \cos(\theta_{z}), \\
 M_{3/2 \ 1/2 \ H/V}^{(\text{BG})(\ell_{\gamma}=1)} \propto \theta_{k} \sin \theta_{z} \qquad M_{3/2 \ 1/2 \ H/V}^{(\text{BG})(\ell_{\gamma}=2)} \propto \theta_{k}^{2} \cos^{2} \frac{\theta_{z}}{2}.$$
(11)

For the case of the  $\ell_{\gamma} = 0$  and a setting of  $\theta_z = 0$ , then both H and V interactions strengths are equal; this is identical to the behavior for E1 and E2 transitions. However, when varying  $\theta_{\tau}$  to  $\pi/2$ , we see a distinct behavior: the value of the horizontally polarized beam does not change while the vertical goes to zero. The behavior for M3 at  $\pi/2$  [see Figs. 3(a), 3(d), and 3(g)] is then similar to that of E2 at  $\pi/4$ , but with interchanged roles of the horizontal and vertical polarizations (see Figs. 3(b), 3(e), and 3(h)). This can be understood by the fact the E and B vectors are orthogonal in a transverse electromagnetic wave. For higher OAM, another interesting feature is observed: the transition strengths are the same for H- and V-polarized beams independent of the alignment angle. This effect is a direct result of the circular dichroism, similar to Section 3. This is exemplified in Fig. 3(d), where we see that for b = 0, both transition strengths coincide.

#### B. Yb<sup>+</sup> Isotopes and E3-Driven Transition

In this subsection, we consider the electric octupole (E3) transition in ytterbium known for ultraprecise optical frequency standards [58]. Ytterbium has several naturally abundant isotopes. This makes this element an interesting species to study the role of nuclear spin and OAM for the features of the excitation strength. Here, we will compare results for ions with a spinless nucleus as in isotope 172 and with I = 1/2 nuclear spin of isotope 171. The transition from the ground state  ${}^{2}S_{1/2} \rightarrow$  ${}^{2}F_{7/2}$  is driven with light near 467 nm. In the case of  ${}^{172}$ Yb<sup>+</sup>, where the nuclear spin is zero, we can use the formalism that has been exploited in the case of  ${}^{40}$ Ca<sup>+</sup>, and  ${}^{40}$ Ar<sup>13+</sup> transitions will be used also for  ${}^{20}$ Ne<sup>5+</sup> in Section 4.C. The coupling scheme to use for  ${}^{171}$ Yb<sup>+</sup> is the one with the coefficients [Eq. (4)] when the proper quantum number for angular momentum is F = J + I, not accounting for electric hyperfine interaction, e.g., [59].

As we can see in Figs. 3(c), 3(f), and 3(i), the behavior at the alignment angle  $\theta_z = \pi/4$  is conceptually similar to the case of E2. The position of the minima and the relative amplitudes of each transition vary slightly. And there is also a dependence of the relative interaction strengths on the spin content of the nucleus. One can see that the ion's response does not change drastically due to the presence of the hyperfine splitting. The correction due to the presence of the nuclear spin is on the order of 10%.

However, a striking and distinctive feature occurs for E3 transitions in the collinear case:  $\theta_z = 0$ . As we show in Fig. 2, now we can drive  $\Delta m = 3$  transitions. These are particularly strong at the center of the beam with  $m_{\gamma} = 3$ , as expected from the full selection rules and shown in Fig. 2(i).

The matrix elements at vortex center, as calculated for the previous transitions are

$$\begin{split} &M_{10H}^{(\text{BG})(\ell_{\gamma}=0)} \propto i(4-11\theta_{k}^{2})(\cos\theta_{z}+15\cos3\theta_{z}), \\ &M_{10V}^{(\text{BG})(\ell_{\gamma}=0)} \propto -i(4-11\theta_{k}^{2})(3+5\cos2\theta_{z}), \\ &M_{10H}^{(\text{BG})(\ell_{\gamma}=1)} \propto -4\theta_{k}(23+20\cos\theta_{z}+45\cos2\theta_{z})\sin\theta_{z}, \\ &M_{10V}^{(\text{BG})(\ell_{\gamma}=1)} \propto -4\theta_{k}(13-20\cos\theta_{z}+15\cos2\theta_{z})\sin\theta_{z}, \\ &M_{10V}^{(\text{BG})(\ell_{\gamma}=2)} \propto i\frac{3}{2}\theta_{k}^{2}(22+7\cos\theta_{z}+10\cos2\theta_{z}+25\cos3\theta_{z}), \\ &M_{10V}^{(\text{BG})(\ell_{\gamma}=2)} \propto i6\theta_{k}^{2}(21-40\cos\theta_{z}+35\cos2\theta_{z})\cos^{2}\frac{\theta_{z}}{2}. \end{split}$$

Hence, the horizontal polarization is completely suppressed when impact parameter  $b \rightarrow 0$  for  $\theta_z = \pi/2$ . Dichroism effects can be seen for  $\ell_{\gamma} > 2$ , and the full amplitude extinction takes place for  $\ell_{\gamma} \ge 5$  in the central region of the beam for both <sup>171</sup>Yb<sup>+</sup> and <sup>172</sup>Yb<sup>+</sup> isotopes.

Finally in Fig. 4, we plot the profiles for varying polarizations at alignment angle  $\theta_z = \pi/4$ . It is remarkable how a small



**Fig. 2.** Transition strength  $\Omega_r$  for different atomic multipolarities and beam types as a function of impact parameter *b*. The beam direction chosen to be collinear with the external static magnetic field:  $\theta_z = 0$ . Under these conditions, conservation of the projection of angular momentum along *z* is conserved ( $\Delta m = \Lambda + \ell_\gamma$ ) for b = 0. We choose for all plots left-circular polarized photons  $\Lambda = +1$ . Columns group multipolarities (*M*1, *E*2, *E*3), while rows group results of increasing orbital angular momentum  $\ell_\gamma = 0, 1, 2$ . Line type indicates the magnetic transition  $\Delta m = 1, 2, 3$  for solid green, dashed–dotted brown, and dotted red. For magnetic dipole transitions, only  $\Delta m = 1$  transitions are present, and there is a maximum at the center only for  $\ell_\gamma = 0$ , where the conservation of the projection of angular momentum is conserved. For electric quadrupole (*E*2) transitions, also  $\Delta m = 2$  is allowed when  $\ell_\gamma = 1$ , as seen in subplot (e). There is a nonzero interaction at the dark center of the beam. Similarly, for the electric octupole (*E*3),  $\Delta m = 3$  transitions are allowed. As in all cases, transitions are allowed for b = 0 only when the conservation rule is met:  $\Delta m = \Lambda + \ell_\gamma$ . All plots for *E*3 transitions also show an expanded inset with the detail of behavior for  $\Delta m = 3$ . In (h), we contrast our predictions to the results presented in Ref. [11].



**Fig. 3.** Transition strength  $\Omega_r$  for different atomic multipolarities and beam types as a function of impact parameter *b*. In all cases, the atomic change in magnetic number is  $\Delta m = 1$ . Columns group multipolarities (*M*1, *E*2, *E*3), while rows group results of increasing orbital angular momentum  $\ell_{\gamma} = 0, 1, 2$ . Red solid (blue dashed) line indicates results for light polarized vertically (horizontally). For *E*3, we also show the corresponding results for non-null nuclear spin (I = 1/2) in dotted purple (dashed–dotted black). We observe the inclusion of nuclear spin in the *E*3 transition produces only a difference in overall strength but no change in structure. Electric transitions (*E*2 and *E*3) are for alignment angle  $\theta_z = \pi/4$ ; they show qualitatively similar behavior. In subplots (b) and (e), we contrast our predictions with experimental data from [11]. The magnetic *M*1 transition is shown at alignment angle  $\theta_z = \pi/2$ , where it also shows qualitatively similar behavior to the electric quadrupole and octupole but with the polarizations inverted.

variation in polarization ( $\Delta \phi_b = \pm 17^\circ$ ) results in strong visual effects of mirror symmetry breaking.

# C. <sup>20</sup>Ne<sup>5+</sup> and E2 + M1 Separation

In this section, we consider  ${}^{2}P_{1/2} \rightarrow {}^{2}D_{3/2}$  transitions in a B-like  ${}^{20}\text{Ne}^{5+}$  HCI at  $\lambda = 142$  nm, where  $M1/E2 \propto 1.1$  [46]. As before, we use the multipole expansion of Eq. (3) to get the regular contributions into the plane wave absorption amplitude as

$$M_{m_f m_i \Lambda}^{(\text{pw})} \propto \sqrt{5} C_{1/2 m_i; 2\Lambda}^{3/2 m_f} E2 + \Lambda \sqrt{3} C_{1/2 m_i; 1\Lambda}^{3/2 m_f} M1.$$
 (13)

With the use of Eqs. (1)–(3), one can work out the explicit form of expressions for the rotated transition amplitudes and predict the Rabi frequencies for this case. In the general case, the norm of the transition amplitude is not always free of interference terms. However, in this case, no such terms arise. For this reason, the behavior for this transition is simply a linear combination of the ones for M1 and E2, as described above and shown in Figs. 3 and 5.

For this HCI, we study the transition amplitudes for horizontal and vertical polarizations, as shown in Fig. 5. The resulting amplitude for a transition with a mixture of M1 and E2 is shown in the first column. The individual multipolar contributions are shown in the second and third columns, E2 and M1, respectively. One can clearly see that the quadrupole contribution (second column) looks identical to the E2 transition discussed above for calcium ions (Fig. 2(b), 2(e), and 2(h)), as well as the magnetic dipole contribution (third column) is identical to M1 transition discussed above for argon HCIs.

Let us examine this behavior mathematically in proximity to the beam vortex in this case. For  $\ell_{\gamma} = 0$  one gets



**Fig. 4.** Contour plots of the normalized transition strength as a function of polarization and impact parameter for  ${}^{2}S_{1/2} \rightarrow {}^{2}F_{7/2}$  in a single  ${}^{172}$ Yb<sup>+</sup>. In both cases,  $\ell_{\gamma} = 1$ ,  $\delta = 0$ ; but  $\phi_{b} = 0$  (-0.3 rad) for left (right) plot.



**Fig. 5.** Transition strength  $\Omega_r$  for different atomic multipolarities and beam types as a function of impact parameter *b*. The left column shows the expected response for a transition with similar contribution from the *M*1 and *E*2 multipoles, such as for the HCl <sup>20</sup>Ne<sup>5+</sup> on the transition <sup>2</sup>P<sub>1/2</sub>  $\rightarrow$  <sup>2</sup>D<sub>3/2</sub> [46]. Columns *E*2 and *M*1 show the individual contribution from each of the multipoles. In all the cases,  $\Delta m = 1$ ,  $\phi_b = 0$ , and the alignment angle is chosen to be  $\theta_z = \pi/4$ . Each row shows the expected result for increasing orbital angular momentum  $\ell_{\gamma} = 0, 1, 2$ . Red solid (blue dashed) line indicates results for light polarized vertically (horizontally) beams.

$$M_{3/2\ 1/2\ H}^{(BG)} \propto i \left(\sqrt{3}M1 - E2(1 - 2\ \cos\ \theta_k)\ \cos\ 2\theta_z\right),$$
  
$$M_{3/2\ 1/2\ V}^{(BG)} \propto i \left(\sqrt{3}M1 - E2(1 - 2\ \cos\ \theta_k)\ \cos\ \theta_z\right).$$
(14)

For  $M1 \rightarrow 0$ , we get the same approximation as in the purely E2-driven case [Eq. (9)]. In the same way,  $E2 \rightarrow 0$  gives Eq. (11). The same can be proven for higher values of OAM  $\ell_{\gamma}$ . This means that for the case with mixed multipolarity, the horizontal component cannot be completely suppressed at any  $\alpha$ , unless magnetic contribution is negligible or the pitch angle  $\theta_k$  is very small. Second, however, would mean taking the limit of OAM going to zero—the plane wave limit. Hence, presence of strong M1 contribution sensibly changes the photo-absorption behavior at the beam center.

Both  ${}^{20}$ Ne<sup>5+</sup> and  ${}^{40}$ Ar<sup>13+</sup> have first nonzero contribution coming from a magnetic dipole. In Figs. 5(a), 5(d), and 5(g), the observed drastic difference in neon optical response in the area of the beam penumbra, compared to argon in Figs. 3(a), 3(d), and 3(g), is due to the presence of the second strongest multipole—the electric quadrupole. It can be inferred that contributions coming from higher order multipoles may result into visible effects in the central region, as in this case, turning minima into maxima in position dependence of Rabi frequency. This would be especially useful for fundamental studies in spectroscopy of HCIs.

# 5. CONCLUSION AND OUTLOOK

In this paper, we have studied the modifications of selection rules for electric and magnetic transitions, particularly E2, E3, and M1, when well-localized single ions interact with

focused tailored electromagnetic fields with vortices. We have developed a complete theoretical framework, extending previous results, e.g., [36], where we can determine the full dependence of the photo-absorption transition amplitudes on OAM, on polarization of the beam, on impact parameter, on nuclear spin when relevant, and on directions of the quantization axis of the atoms. This whole framework for single-ion–vortex interactions is tested and verified using experimental data for  $^{40}Ca^+$  [11].

We also encourage development of future experiments involving twisted photon photo-absorption on various ions, including Yb (used for clocks) and other HCIs, by predicting rates for selected transitions and a variety of structured electromagnetic beams. We highlight peculiar effects, which can form a rich ground of fundamental studies, and will, e.g., allow separating the E and M character for given atomic transitions and enhance the excitation rates in certain circumstances.

The calculations give transition amplitudes, which for fixed laser strength are equivalent to Rabi oscillation frequencies, as experimentally measured. The experimental results for the <sup>40</sup>Ca ion, which confirmed the validity of the calculations, were for the case with a 45° static magnetic field alignment and in the full polarization domain of the laser beam for OAM  $\ell_{\gamma} = 0$  and 1. Together with the predictions in related configurations for Ar, Ne, and Yb ions, one may conclude that local, or small impact parameter, alteration of the ordinary spectroscopic selection rules enables selective enhancement of high-order multipolar contributions in the ion response by manipulating the photon twist. Circular dichroism related to the twisted photon topology is theoretically seen on the level of factorized multipolar contributions. The reported circular dichroism in

$\Delta m$			
H/V	0	±1	±2
Н	$\sin(2\theta_k)\cos\psi_k$	$\pm \cos 2\theta_k \cos \psi_k - i \cos \theta_k \sin \psi_k$	$-\frac{1}{2}\sin(2\theta_k)\cos\psi_k\pm i\sin\theta_k\sin\psi_k$
V	$i \sin(2\theta_k) \sin \psi_k$	$-\cos\theta_k\cos\psi_k \pm i\cos(2\theta_k)\sin\psi_k$	$\pm \sin \theta_k \cos \psi_k - i \frac{1}{2} \sin(2\theta_k) \sin \psi_k$
Н	$-\sin(2\theta_k)$	$\pm \cos(2\hat{\theta}_k) \cos \hat{\phi}_k + i \cos(2\hat{\theta}_k) \sin \hat{\phi}_k$	$\sin(2\theta_k)\cos(2\phi_k) \pm i\sin(2\theta_k)\sin(2\phi_k)$
V	0	$-\cos \theta_k (\cos \phi_k \pm i \sin \phi_k)$	$-\sin \theta_k(\pm \cos(2\phi_k) + i\sin(2\phi_k))$

Table 1. Geometry-Dependent Terms in the Plane Wave Photo-Absorption Matrices [Eq. (A1)] for Two Sets of Euler Angles: Top,  $\{\psi_k, \theta_k, 0\}$ ; Bottom,  $\{0, -\theta_k, -\phi_k\}$ 

atom-photon interactions may contribute into the study of chiral light-matter interactions [60].

The ions and transitions studied in this paper are excited by different multipolarities. The results from horizontal and vertical linear polarization states become most distinct at particular static magnetic field alignment angles  $\theta_z$ , with the angle of most sensitivity depending on the photon-atom angular momentum transfer. For the system with M1 transitions, or partial wave amplitude with j = 1, choose  $\theta_z = \pi/2$ ; for E2 or j = 2, choose  $\theta_z = \pi/4$ ; and for E3 or j = 3, choose again  $\theta_z = \pi/2$ . For the Boron-like <sup>20</sup>Ne<sup>5+</sup> HCI, the M1 and E2 partial waves are unusually similar, leading to effects shown in this paper (see also [32]). This makes this HCI a promising candidate for future experimental research of topological effects in lightmatter interactions. Progress in this direction will also allow further investigation of the quantum nature of spin-orbit coupling in photon laser beams.

Also included are a number of results for  $\theta_z = 0$ , with transition amplitudes for differing OAM  $\ell_{\gamma}$  with positive circular polarization  $\Lambda$ , and with differing changes in the atomic angular momentum projections,  $\Delta m$ . One sees again [11,36] the nonzero response at the zero impact parameter only for  $\Delta m = m_{\gamma} = \ell_{\gamma} + \Lambda$ , and of note is the good agreement with experimental data available in a range of impact parameters for one of the <sup>40</sup>Ca<sup>+</sup> cases.

Photon OAM coupling to the internal atomic degrees of freedom can be used, as seen here, to separate the electric and magnetic character of given transitions, to locally enhance next-to-leading order transition rates, and to suppress parasitic transitions. In the realm of quantum computation, incorporation of OAM states of light can provide ion-photon interaction on the level of energy, momentum, polarization, and phase.

The theory outlined in this paper can also be successfully applied to such systems as bulk semiconductors and artificial atoms in the ways similar to [61,62]. When developed, it may become useful in such fields as metrology, classical and quantum communication, quantum computing, high-capacity data transfer, and cybersecurity, to name a few. The highlighted sensitivity to the target position in the beam, polarization, and phase can be used in beam diagnostics.

# **APPENDIX A**

One might benefit from linking two alternative sets of angles  $\{\pm\psi_k, \pm\theta_k, \pm\phi_k\}$ , equivalent to active and passive 3D rotation. The conventional formalism [32,63] for photo-absorption of the plane wave of arbitrary incidence angle  $\Omega$  by the atomic system may be written as

$$M_{m_{f}m_{i}\Lambda}^{(\text{pw})}(\Omega) = -\sqrt{4\pi} \sum_{j=1}^{\infty} \sum_{m=-j}^{j} i^{j+\mu} \sqrt{\frac{2j+1}{2j_{f}+1}} \Lambda^{\mu+1} \times D_{\Lambda m}^{j*}(\psi_{k},\theta_{k},0)(-1)^{j-j_{f}+j_{i}} C_{i,m,im}^{jfm_{f}} M_{i\mu},$$
(A1)

where the rotation matrix is related to ours as  $D^{j}_{mm'}(\psi_k, \theta_k, \phi_k) = e^{-im\psi_k} d^{j}_{mm'}(\theta_k) e^{-im'\phi_k}$ . This matrix represents active Euler rotation in Hilbert space. The part of the tensor responsible for the spatial configuration of the system,

$$\Lambda D_{\Lambda m}^{j*}(\psi_k, \theta_k, 0) C_{j_j m_j m}^{j_f m_f}, \qquad (A2)$$

reproduces up to a sign the one in semiclassical formalism [53,54]. It can be checked in direct calculation, and the results are presented in the top part of Table 1.

If, instead of rotating the photon state, we rotate an electron state, similar to Eq. (1), then

$$M_{m_{f}m_{i}\Lambda}^{(\text{pw})}(\Omega) = -\sqrt{4\pi} \sum_{j=1}^{\infty} \sum_{m'_{f},m'_{i}} i^{j+\mu} \sqrt{\frac{2j+1}{2j_{f}+1}} \Lambda^{\mu+1} \\ \times D_{m_{f}m'_{f}}^{j_{f}}(0,-\theta_{k},-\psi_{k}) \\ \times D_{m_{i}m'_{i}}^{j_{i}*}(0,-\theta_{k},-\psi_{k})(-1)^{j-j_{f}+j_{i}} C_{j_{i}m'_{j}\Lambda}^{j_{f}m'_{f}} M_{j\mu}.$$
 (A3)

This is the passive rotation  $\{0, -\theta_k, -\psi_k\}$ , equivalent to  $\{\psi_k, \theta_k, 0\}$ . The configuration-dependent part of this equation,

$$\sum_{m'_{f}, m'_{i}} \Lambda C^{j_{f}m'_{f}}_{j_{i}m'_{j}j\Lambda} D^{j_{f}}_{m_{f}m'_{f}}(0, -\theta_{k}, -\psi_{k}) D^{j_{i}*}_{m_{i}m'_{i}}(0, -\theta_{k}, -\psi_{k}),$$
(A4)

is identical to Eq. (A2), though containing an extra Wigner rotation matrix. In our formalism, Section 2, we have used the rotation system, defined in terms of the Euler angles in Eq. (A3) as  $\{0, -\theta_k, -\phi_k\}$ . The resulting geometry-related terms are presented in the bottom half of Table 1. If we proceed with this alternative description in terms of active rotation, we get the equation, analogous to Eq. (1),

$$M_{m_f m_i \Lambda}^{(\text{BB})}(b; \theta_z = 0) = A \sum_m i^{m-2m_\gamma} e^{i(m_\gamma - m)\phi_k} J_{m_\gamma - m}(\kappa b)$$
$$\times d_{m\Lambda}^j(\theta_k) M_{m_f m_i \Lambda}^{(\text{pw})}(0), \qquad \text{(A5)}$$

where the plane wave amplitude is defined as Eq. (3) with the coupling coefficient being

$$C_{jj_f j_i}^{\Lambda} = (-1)^{j_{-j_f} + j_i} C_{j_i, m_i, j, m}^{j_f, m_f}.$$
(A6)

As one can see, this way photon TAM projection  $m_{\gamma}$  explicitly dictates multipolar balance in the optical response of the ion in the beam penumbra, as discussed in Ref. [55].

# **Research Article**

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