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Photoexcitation of atoms near the center of vortex light

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ABSTRACT

We investigate the interaction of atoms with a Laguerre-Gaussian beam near the vortex center where an annular nodal area exists on and around the beam propagation axis. To grasp the essential features related to the orbital angular momentum transfer to a bound electron, we first show that it suffices to include in the interaction Hamiltonian the leading (second) term in the usual expansion of $e^{-i\mathbf{k}.\mathbf{r}}$ to be associated with the transverse (longitudinal) field component of a beam with orbital angular momentum $\ell = \pm 1$. We compare the obtained results for the quadrupole transition $4S_{1/2}$ to $3D_{5/2}$ in Ca^+ with published experimental data. For orbital angular momenta $|\ell| > 1$, this approach predicts that quadrupole transitions are only possible for the configuration where the orbital angular momentum with $|\ell| = 2$ is antiparallel to the spin degree of freedom. These results are confirmed by another approach based on the multipolar Hamiltonian which, additionally, provides rich information on the emergence of different selection rules for arbitrary value of ℓ including their spatial dependence in the region of interest. This work also extends the discussion of the excitation of target atoms by twisted light to two-electron atomic systems by deriving selection rules for single-electron transitions.

There has been renewed and growing interest in fundamental light–atom interaction processes due to the emergence of new possibilities of interaction configurations arising from the advances in laser technology and atom trapping and cooling techniques [1,2]. In particular, light fields can be designed with a well-defined projection of the orbital angular momentum (OAM) onto their propagation axis. These spatially structured light beams, known as vortex beams or twisted light, have stimulated numerous and diverse new applications in imaging, communications and quantum information [3,4]. Atomic photoexcitation by twisted light has been the subject of many theoretical [5–11] and experimental studies [12–15] which were mostly limited to hydrogen-like systems.

Atom–light interaction is commonly described using two types of Hamiltonians. The standard approach employs the minimal-coupling Hamiltonian while the other one uses the multipolar Hamiltonian. In principle and in the absence of approximations, both Hamiltonians are equivalent and related to each other by a unitary transformation; the Power–Zienau–Wolley transformation [16,17].

This paper presents a comparative study of different approaches based on these types of Hamiltonians to derive the angular-momentum quantum selection rules for the twisted-light-photoexcitation of a bound electron. In particular, we focus on the special case where the atom is close the vortex core of a Laguerre-Gaussian beam. The importance of understanding twisted-light–atom interaction near the central point stems from the presence of a phase singularity at the core which is a defining property of twisted light beams.

Laguerre Gaussian (LG_p^ℓ) beams [18] are solutions of the paraxial Helmholtz equation with a cylindrical symmetry and are characterized by two indices ℓ and p. The first index indicates the angular momentum number while the second one corresponds to the number of radial nodes. The vector potential $\mathbf{A}_{\ell p}$ for such circularly polarized beams can be written in the following form:.

$$\mathbf{A}_{\ell p} = \frac{(\hat{x} + i\sigma\hat{y})}{\sqrt{2}} A_0\, u(\mathbf{r}) v(\mathbf{r})\, e^{(-i\omega t + ikz)} + c.c, \tag{1}$$

where $\sigma = 1(-1)$ stands for left (right) circular polarization and $u(\mathbf{r})$ describes the part of the mode function related to the orbital angular momentum:

$$u(\mathbf{r}) = \sqrt{\frac{p!}{(|\ell| + p)!}} \left(\frac{\sqrt{2}\rho}{w(z)}\right)^{|\ell|} \mathcal{L}_p^{|\ell|}\left(\frac{2\rho^2}{w^2(z)}\right) \times \exp\left(il\varphi - i(2p + |\ell|)\arctan(\frac{z}{z_R})\right) \tag{2}$$

while $v(\mathbf{r})$ is the part of the mode function related to the regular Gaussian beam :

$$v(\mathbf{r}) = \frac{w_0}{w(z)} \exp\left(\frac{-\rho^2}{w(z)^2} + \frac{ik\rho^2}{2R(z)^2} - i\arctan(\frac{z}{z_R})\right) \tag{3}$$

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where z is the axial distance from the beam's focus (or "waist"), ρ is the radial distance from the center axis of the beam, A_0 the vector potential at the origin ($\rho = 0, z = 0$), $w(z)$ is the radius at which the field amplitudes fall to $1/e$ of their axial values, $w_0 = w(0)$ is a waist radius, $\mathcal{L}_p^{|\ell|}(x)$ is an associated Laguerre polynomial, z_R is the Rayleigh length, and $R(z)$ is the radius of curvature of the beam's wavefronts at z.

The current study considers beams with $p = 0$ and the case where the atomic wavepacket is located in the vicinity of the optic axis with its size assumed small compared to the lateral spatial extent of the beam. Therefor, the spatial variations due to $v(\mathbf{r})$ can be ignored [9].

Using $\mathbf{A}_{\ell 0}$ and the scalar potential Φ obtained from the Lorenz condition ($\nabla.\mathbf{A} = -\frac{1}{c^2}\frac{\partial \Phi}{\partial t}$), the positive-frequency part of the electric field $\mathbf{E}^+$ of a circularly polarized LG_0^{ℓ} beam can be derived from the mode function as follows:

$$\begin{aligned}\mathbf{E}^{(+)}(\mathbf{r}) = \frac{E_0}{\sqrt{2}} e^{ikz}[\,(\hat{x} + i\sigma\hat{y})\,u(\mathbf{r}) \\ + \hat{z}\,\frac{i}{k}\,(\frac{\partial u(\mathbf{r})}{\partial x} + i\sigma\frac{\partial u(\mathbf{r})}{\partial y})]\end{aligned} \tag{4}$$

where $E_0 = -i\omega A_0$ is related to the average power of the beam P by the relation [19]: $P = (\pi\epsilon_0 c\, w_0^2/4)\,|E_0|^2$. Here we ignore the derivatives of the scalar potential with respect to the transverse coordinates which are beyond the validity of the paraxial approximation [20].

The expressions of the transverse and longitudinal components of $\mathbf{E}^+$ close to the phase singularity at $z = 0$ can be simplified [9], without considerable loss of accuracy, into:

$$\mathbf{E}_{\perp}^{(+)}(\mathbf{r}) = \frac{(\hat{x} + i\sigma\hat{y})}{\sqrt{2}}\,\frac{E_0}{\sqrt{|\ell|!}}\left(\frac{\sqrt{2}}{w_0}\rho\right)^{|\ell|} e^{i\ell\varphi} e^{ikz} \tag{5}$$

$$\begin{aligned}\mathbf{E}_{\mathbf{z}}^{(+)}(\mathbf{r}) = -\,i\,\hat{z}\,(l\sigma - |\ell|)\,(1 - \delta_{\ell,0})\,\frac{E_0}{\sqrt{|\ell|!}}\,\frac{1}{w_0 k} \\ \times\left(\frac{\sqrt{2}}{w_0}\rho\right)^{|\ell|-1} e^{i(\ell+\sigma)\varphi} e^{ikz}\end{aligned} \tag{6}$$

For $\ell = 0$, the approximated field expression for a Gaussian beam can be obtained.

The minimal-coupling Hamiltonian for an electron in the field reads:

$$H = \frac{1}{2m}(\mathbf{p} + e\mathbf{A})^2 + V(\mathbf{r}) - e\Phi \tag{7}$$

In the following part, we will derive the selection rules based on this Hamiltonian using the Coulomb gauge:

$$H = \frac{1}{2m}\mathbf{p}^2 + V(\mathbf{r}) + \frac{e}{m}\mathbf{A}^{\mathbf{C}}.\mathbf{p} = H_0 + \frac{e}{m}\mathbf{A}^{\mathbf{C}}.\mathbf{p} \tag{8}$$

where the quadratic term of $\mathbf{A}^{\mathbf{C}}$ (the vector potential expressed in Coulomb gauge) is ignored and H_0 represent the atomic Hamiltonian.

Next, we make the usual expansion of the term (e^{ikz}) in the field expression. In contrast to [21], we consider both the first and second terms in the expansion to study quadrupole transitions. The matrix element of the interaction part of the Hamiltonian is then:

$$\begin{aligned}\frac{i\,e}{m\omega}\,\langle f|\mathbf{E}.\mathbf{p}|i\rangle &= \frac{i\,e}{m\omega}\,\langle f|(\mathbf{E}_{\perp}^{+} + \mathbf{E}_{\mathbf{z}}^{+})(1 + ikz)\,.\mathbf{p}|i\rangle \\ &= \frac{i\,e}{m\omega}\,\langle f|E_z^{+}\,p_z + E_{\perp}^{+}\,(p_x + i\sigma p_y) \\ &\quad + E_z^{+}\,(ikz)\,p_z + ..|i\rangle\end{aligned} \tag{9}$$

where $\mathbf{E}_{\perp}^{+}$ ($\mathbf{E}_{\mathbf{z}}^{+}$) stands for $\mathbf{E}_{\perp}^{+}$ ($\mathbf{E}_{\mathbf{z}}^{+}$) without ($e^{ikz}$). Based on this particular formalism, it is easier to consider first the case of a beam with $\ell = \pm 1$.

The first term in Eq. (9) can be written using the usual dipole approximation in near-resonance regime as:

$$\frac{i\,e}{m\omega}\langle f|E_z^{+}\,p_z|i\rangle = e\,\langle f|E_z^{+}\,z|i\rangle \tag{10}$$

where the commutation relation: $[\mathbf{r}, H_0] = \frac{i\hbar}{m}\,\mathbf{p}$ is used.

Writing the expression ($\rho e^{i\ell\phi} = x + i\,\ell\,y$) in Cartesian coordinates, the second and the third terms contain expressions of the form: $\langle f|r_m\,p_n|i\rangle$ with $m, n = 1, 2, 3$ where r_m and p_n denote the component of the vector operators for position and momentum, respectively. It is convenient to write these forms as:

$$\langle f|r_m\,p_n i\rangle = \frac{1}{2}\,(\langle f|r_m\,p_n - p_m\,r_n|i\rangle + \langle f|r_m\,p_n + p_m\,r_n|i\rangle) \tag{11}$$

In contrast to the antisymmetric combination, the symmetric one leads to quadrupole transitions [22] and can be simplified using the above commutation relation to:

$$\frac{1}{2}\,\langle f|r_m\,p_n + p_m\,r_n|i\rangle = -\frac{i\hbar\,m\omega_{fi}}{2}\,\langle f|r_m\,r_n|i\rangle \tag{12}$$

Grouping these terms and transforming them into spherical coordinates, one can construct the part of the Hamiltonian responsible for quadrupole transitions H_{dq}. The transition amplitude due to H_{dq} is then:

$$\begin{aligned}\langle f|H_{qd}|i\rangle = -\,\frac{\alpha}{2}\,\langle f|r^2\sin^2(\theta)e^{i(\ell+\sigma)\varphi}|i\rangle \\ + \delta_{\ell,-\sigma}\,\alpha\,\langle f|r^2\cos^2(\theta)|i\rangle\end{aligned} \tag{13}$$

where $\alpha = eE_0/w_0$. Writing the interaction matrix in terms of spherical tensors, one obtains:

$$\begin{aligned}\ell = \sigma = \pm 1 :\\ H_{qd} = -\sqrt{\frac{8\pi}{15}}\alpha r^2\,T_{\pm 2}^{(2)} \\ \ell = -\sigma :\\ H_{qd} = \sqrt{\frac{3}{2}}\sqrt{\frac{8\pi}{15}}\alpha r^2\,T_0^{(2)}\end{aligned} \tag{14}$$

According to the expression (9), there is no electric dipole interaction term for a beam with orbital angular momentum greater than $1\hbar$. Moreover, only the longitudinal part of the electric field gives rise to an electric quadrupole transition for ($|\ell| = 2$) beams, where the orbital and spin angular momenta are antiparallel to each other. The amplitude of this transition which satisfies the selection rule $\Delta m = \ell + \sigma$ is then:

$$\begin{aligned}\langle f|H_{qd}|i\rangle &= -\frac{4i\,\alpha}{w_0\,k}\,\langle f|r^2\sin(\theta)\cos(\theta)\,e^{i(\ell/2)\varphi}|i\rangle \\ &= \frac{\ell}{2}\,\frac{2i\,\alpha}{w_0\,k}\,\sqrt{\frac{8\pi}{15}}\,\langle f|r^2\,T_{\ell+\sigma}^2|i\rangle\end{aligned} \tag{15}$$

In comparison with the case of $|\ell| = 1$ shown in (14), the probability of this transition is much smaller given the scaling factor of $(w_0 k)^{-1}$. Other than this particular situation, there is no quadrupole transition for cases with $|l| > 1$ within the frame of the above treatment near the singularity point ($r \to 0$).

At this stage, it is important to compare the main results with an archetypal case which has been investigated experimentally. We, therefore, consider a quadrupole transition from the $4S_{1/2}$ to $3D_{5/2}$ in $^{40}Ca^+$ and calculate the relevant matrix elements using the Wigner–Eckart theorem. The resultant calculations summarized in Table 1 show the dependence of the quadrupole transition on the sign of the orbital angular momentum of the photon. The general trend of the theoretical relative transition strength follows that of the relative experimental power-normalized Rabi frequencies [15], which are proportional to the matrix elements. The agreement is excellent for the ratios of the LG beams while it is less satisfactory for those of Gaussian beams which is probably due to experimental difficulties associated with changing the type of the beams [9].

It is interesting to expand the same treatment to OAM transfer to two-electron atoms. In particular, we consider the photo-excitation by an $\mathrm{LG}_0^{\pm 1}$ beam from the following two-electron ground state:

$$|\psi_i(r_1, r_2)\rangle = |\psi_{1s}(r_1)\rangle|\psi_{1s}(r_2)\rangle \tag{16}$$

to a final singly excited state (S=0) given as:

$$|\psi_f(r_1, r_2)\rangle = \frac{1}{\sqrt{2}}(|\psi_{1s}(r_1)\rangle|\psi_{3d}(r_2)\rangle + |\psi_{3d}(r_1)\rangle|\psi_{1s}(r_2)\rangle) \tag{17}$$

Table 1
Calculated normalized quadrupole transition matrix element for the transition $4S_{1/2}$ to $3D_{5/2}$ in Ca^+ with $m_i = -\frac{1}{2}(+\frac{1}{2})$. Normalization is done with respect to the $\Delta m = -2(+2)$ transition. Corresponding experimental ratios obtained from [15] are in brackets. The beam parameters are of $\lambda = 729$ nm and $w_0 = 2.7$ μm.

Δm	l, σ	m_i=−1/2	m_i=1/2
−2	−1,−1	1 (1)	0.45 (0.48)
−1	0,−1	7.32 (10.69)	5.20 (8.62)
0	1,−1/ −1,1	0.95 (0.95)	0.95 (1.04–0.94)
1	0,1	5.20 (6.58)	7.32 (12.3)
2	1,1	0.45 (0.43)	1 (1)

The transition amplitude $\langle\psi_f(r_1,r_2)|H|\psi_i(r_1,r_2)\rangle$ can be evaluated using the Wigner–Eckart theorem for the initial state with J=m_j=0 and the final state with J=2. The coupling between these two states is nonzero only for tensors of rank 2. The evaluation of the related expressions indicates that transitions occur when the difference in angular momentum projection Δm between initial and final atomic states equals to the total angular momentum of the exciting photon. The relative transition strengths for $\Delta m = \pm 2, \Delta m = \pm 1$ and $\Delta m = 0$, are: 1, $\pi w_0/(\sqrt{2}\lambda)$ and $\sqrt{3/2}$, respectively. This particular case shows that the strength of twisted light absorption in the target is insensitive to the sign of the optical angular momentum. On the other hand, this symmetry is removed for photo-excitation from a singly-excited state to a higher one, such as from ^{1}P to ^{1}F.

Quinteiro et al. [9,23] interpreted the experimental data of [15] starting from the Hamiltonian in Eq. (7) and using the Poincare gauge [24] where the potentials are defined as:

$$\begin{aligned}\mathbf{A^P}(\mathbf{r},t) &= -\int_0^1 d\mu\, \mu\mathbf{r}\times\mathbf{B}(\mu\mathbf{r},t)\\ \Phi^P(\mathbf{r},t) &= -\int_0^1 d\mu\, \mathbf{r}\cdot\mathbf{E}(\mu\mathbf{r},t).\end{aligned} \tag{18}$$

In this case, the interaction Hamiltonian comes only from the scalar potential. i.e $H_{qd} = e\Phi^P(\mathbf{r},t)$. In spherical coordinates, the interaction Hamiltonian separated into transverse and longitudinal parts reads for $\ell = \pm 1$:

$$\begin{aligned}H_\perp^{(+)} &= -\alpha r^2\sin^2(\theta)e^{i|\ell|/(\ell+\sigma)}\cdot\int_0^1 \mu e^{ik\mu\rho\cos(\theta)}d\mu\\ H_z^{(+)} &= -i\delta_{\ell,\sigma}\alpha r\cos(\theta)\frac{2}{k}\int_0^1 e^{ik\mu\rho\cos(\theta)}d\mu.\end{aligned} \tag{19}$$

In order to apply the approximations at the vortex core consistently, the exponentials in (19) are expanded and the zeroth order is kept in the transverse part of the Hamiltonian while the first order is kept in the longitudinal part. Performing the integration and writing the Hamiltonians in terms of spherical tensors, one can obtain the same spherical tensor components as in Eq. (14).

Although the calculations in Ref. [9] lead to the same ratios presented in Table 1 for the selection rule: $\Delta m = \ell + \sigma$ with $\ell = \pm 1$, they differ in some details. First, the Hamiltonian for the transition driven by an LG beam with ($\ell = \pm 1$ and $\sigma = -\ell$) involves a tensor of rank 1. This implies that this term multiplied by a radial integral of r^2 leads to an electric-dipole type transition with a strength comparable to the quadrupole transition. The incorrect appearance of the dipole-type term in [9] comes from the step of converting the product of spherical tensors of rank 1 to a sum of tensors of rank 2. If one uses the correct form of the conversion formula [25] such term will not appear and complete agreement with Eq. (14) can be obtained. In fact, dipole transitions are allowed by the matrix element (10), which associates the longitudinal component of the field $\mathbf{E}_\mathbf{z}^{(+)}$ with the leading term in the expansion of e^{ikz}. Giammanco et al. [14] showed experimentally that there is no observable effect of the OAM on the electric-dipole selection rules. Nevertheless, the dipole interaction due to the OAM-beam in the described case is unnoticeable given the experimental setup of [14] since the factor $(kw_0)^{-1}$ appears as a scaling factor [26]. As an additional difference, the transitions driven by the Gaussian beam ($l = 0, \sigma = \pm 1$) presented in Table 1 are calculated, by taking all coefficients into account, to be weaker by a factor of $\sqrt{2}$ compared to the ones listed in Ref. [9].

For the sake of completing the discussion, it would be fruitful to investigate how the Power–Zienau–Woolley (PZW) formalism [6,16,17] is not only capable of confirming the previous results of the selection rule $\Delta m = \ell + \sigma$ but also provides information on the emergence of different selection rules for any value of ℓ taking into their spatial dependence in an elegantly compact form. In this formalism, the coupling of the atom to the electric field is written in the form of a multipolar series expansion about the center of mass coordinates $\mathbf{R} = (X, Y, Z)$ as follows:

$$H_{int} = H_{dp} + H_{qd} + \cdots. \tag{20}$$

where H_{dp} and H_{qd} represent the electric dipole and quadrupole interactions, respectively. The term H_{qd} is responsible for the relevant quadrupole transition and given by:

$$H_{qd} = -\frac{1}{2}\sum_{ij} Q_{ij}\nabla_i E_j \tag{21}$$

where $Q_{ij} = e\, x_i x_j$ stands for the quadrupole transition operator and x_i are the components of the internal position vector. It is worth pointing out that the electric field vector is expressed here as a function of the center of mass position vector $\mathbf{R}$.

The matrix elements for atomic excitation by an LG_0^ℓ beam can be, therefore, expanded as:

$$\begin{aligned}\langle f|H_{qd}|i\rangle &= -\frac{1}{2}\sum_{ij}\langle f|Q_{ij}|i\rangle\nabla_i E_j\\ &= -\frac{1}{2}\langle f|(Q_{xx} + i\sigma Q_{xy})|i\rangle\frac{E_X}{R\sin(\theta)}M\\ &\quad -\frac{1}{2}\langle f|(Q_{yx} + i\sigma Q_{yy})|i\rangle\frac{E_X}{R\sin(\theta)}N\\ &\quad -\frac{1}{2}\langle f|(Q_{zx} + i\sigma Q_{zy})|i\rangle E_X(ik)\\ &\quad -\frac{1}{2}[O\langle f|Q_{xz}|i\rangle + P\langle f|Q_{xz}|i\rangle\\ &\qquad +(ikR\sin(\theta))\langle f|Q_{zz}|i\rangle]\frac{E_Z}{R\sin(\theta)}\end{aligned} \tag{22}$$

where E_X, $E_Y = i\sigma E_X$ and E_Z are the electric field component along X, Y and Z direction.

Writing the interaction matrix in terms of spherical tensors, we have:

$$\begin{aligned}\langle f|H_{qd}|i\rangle &= -\sqrt{\frac{\pi}{30}}e[(M-iN)(1+\sigma)\frac{E_X}{R\sin(\theta)}\langle f|r^2T_2^2|i\rangle]\\ &\quad +(M+iN)(1-\sigma)\frac{E_X}{R\sin(\theta)}\langle f|r^2T_{-2}^2|i\rangle\\ &\quad -(ik(1+\sigma)E_X + (O-iP)\frac{E_Z}{R\sin(\theta)})\\ &\qquad\times\langle f|r^2T_1^2|i\rangle\\ &\quad +(ik(1-\sigma)E_X + (O+iP)\frac{E_Z}{R\sin(\theta)})\\ &\qquad\times\langle f|r^2T_{-1}^2|i\rangle\\ &\quad +\sqrt{\frac{2}{3}}(-(M+i\sigma N)\frac{E_X}{R\sin(\theta)} + 2ikE_Z)\\ &\qquad\times\langle f|r^2T_0^2|i\rangle\end{aligned} \tag{23}$$

where the functions M, N, O and P are as follows:

$$\begin{aligned}M &= |\ell|\cos(\varphi) - i\ell\sin(\varphi)\\ N &= |\ell|\sin(\varphi) + i\ell\cos(\varphi)\\ O &= (|\ell|-1)\cos(\varphi) - i(\ell+\sigma)\sin(\varphi)\\ P &= (|\ell|-1)\sin(\varphi) + i(\ell+\sigma)\cos(\varphi)\end{aligned} \tag{24}$$

Now we first work out the cases for $\ell = \pm 1$ where the matrix element can be expressed as:

$$\begin{aligned} \ell = \sigma = \pm 1 :& \\ \langle f|H_{qd}|i\rangle = &-\sqrt{\frac{8\pi}{15}}\,[\,\alpha\, e^{ikZ}\,\langle f|r^2\,T^2_{\pm 2}|i\rangle \\ &-\sigma\frac{ik\,E_X}{2}\,\langle f|r^2\,T^2_{\sigma}|i\rangle\,] \\ \ell = -\sigma = \pm 1 :& \\ \langle f|H_{qd}|i\rangle = &\sqrt{\frac{8\pi}{15}}\,[\,\sqrt{\frac{3}{2}}\,\alpha\, e^{ikZ}\,\langle f|r^2\,T^2_{0}|i\rangle \\ &+\sigma\frac{ik\,E_X}{2}\,\langle f|r^2\,T^2_{\sigma}|i\rangle\,] \end{aligned} \tag{25}$$

It is clear that this method also results in the same ratios for the selection rule: $\Delta m = \ell + \sigma$ obtained from Eq. (14). However, there are extra terms proportional to E_X resulting in transitions to states with $\Delta m = \pm 1$. These extra terms do not appear in the previous two approaches because they are accompanied by approximations. It should be noted that on-axis the phase factor $\exp(ikZ)$ will become 1 and E_X will vanish and full agreement with the Hamiltonian expression in (14) will be restored. According to (25), there are two channels for exciting a quadrupole transition in an atomic target positioned in the dark region near the vortex center of an $LG_0^{\pm 1}$ beam. The main channel fulfills the selection rule $\Delta m = \ell + \sigma$ while the weaker channel obeys the rule $\Delta m = \sigma$. In the experiment of Schmiegelow et al. [15] where the ion is excited at the center of a vortex field, the average ratio between the two channels was measurable due to position fluctuations and was in the range ~ 0.28. As the atom moves away from the dark region, the rate of $\Delta m = \sigma$ transitions grows rapidly following the beam profile as confirmed by the experimental work described in [27].

Since $E_X \propto (R\sin(\theta))^{|\ell|}$ and according to Eq. (23), there will be no quadrupole transition due to the transverse component of the field when $R \to 0$ for beams with $|\ell| > 1$. On the other hand, the longitudinal component of the field, which is $E_Z \propto (R\sin(\theta))^{(|\ell|-1)}$, can give rise to transitions only for cases with $|\ell| = 1, 2$. This perfectly confirms the results obtained by the earlier approach in Eqs. (14) and (15). Moreover, Eq. (23) demonstrates the rise of different selection rules for different ℓ values as R increases from zero.

We have compared three different approaches employed to study twisted light absorption by a bound electron and to derive the relevant selection rules in the vicinity of the beam axis. Similarities and differences were highlighted and the outcomes were validated by comparing to a comprehensive set of experimental data. In particular, all approaches result in the same ratios for the selection rule: $\Delta m = \ell + \sigma$ for $|l| = 1$. For LG beams with higher values of the orbital angular momentum, quadrupole transitions are only possible for the case with $|\ell| = 2$ due to the longitudinal field component. The last approach does not involve any approximations and has the advantage of being explicitly sensitive to the spatial variation. For example, it can be used to demonstrate the rise of another selection rule, $\Delta m = \sigma$, whose contribution is zero at the singularity point and becomes progressively significant as the photoexcitation region is moved away from this point. We also have attempted a first extension of these selection rules to two-electron singlet states. The multipolar-Hamiltonian-based approach turned out to be particularly rich and promising for generalization. For example, it should not be difficult to use the multipolar interaction Hamiltonian to treat the more general case taking into account the full spatial dependence of the electric field and different values of ℓ and p. In this case, one needs to re-calculate $\nabla_i E_j$ and subsequently the functions: M, N, O and P in order to follow the changes in off-axis selection rules and transition amplitudes. On the other hand, a more twisted path has to be followed when the calculations are based on the minimal-coupling Hamiltonian For example, one method involves calculating the transition amplitudes due to vortex beams after first expressing them as a sum of plane wave components [8,27,28]. Comparing the different approaches in this context and identifying their advantages and limitations for different beam configurations and target systems are interesting to explore in future work.

CRediT authorship contribution statement

A.F. Alharbi: Developed the idea of the work, Performed the analytic calculations and wrote original draft. **A. Lyras:** Verified the analytical work, Suggested the inclusion of twisted-light interaction with two-electron atomic systems in the work, Contributed to improving the manuscript. All authors discussed the results and contributed to the final manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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