## Photoexcitation of atoms by cylindrically polarized Laguerre-Gaussian beams

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We analyze the photoexcitation of atoms with a single valence electron by cylindrically polarized Laguerre-Gaussian beams. Theoretical analysis is performed within the framework of first-order perturbation theory and by expanding the vector potential of the Laguerre-Gaussian beam in terms of its multipole components. For cylindrically polarized Laguerre-Gaussian beams, we show that the (magnetic) sub-components of electric-quadrupole field vary significantly in the beam cross section with beam waist and radial distance from the beam axis. We discuss the influence of varying magnetic multipole components in the beam cross section on the sublevel population of a localized atomic target. In addition, we calculate the total excitation rate of electric-quadrupole transition ( $4s \ ^2S_{1/2} \rightarrow 3d \ ^2D_{5/2}$ ) in a mesoscopic target of a Ca<sup>+</sup> ion. These calculations shows that the total rate of excitation is sensitive to the beam waist and the distance between the center of the target and the beam axis. However, the excitation by a cylindrically polarized Laguerre-Gaussian beam is found more efficient in driving electric-quadrupole transition in the mesoscopic atomic target than the circularly polarized beams.

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## I. INTRODUCTION

The pioneering work of Allen and co-workers [1] in 1992 showed that the twisted light beam possesses orbital angular momentum (OAM). Since then, research on twisted light (or vortex) beams has been a burgeoning research area in the scientific community [2–5]. The existence of an extra angular momentum in the vortex beam is associated with an azimuthally varying phase of the beam [6]. The presence of OAM in the twisted light beams makes it very suitable for a number of applications such as high harmonic generation [7] and optical tweezers [8]. Moreover, twisted light beams have been utilized as alphabets to encode information beyond one bit per single photon [9]. This makes the Laguerre-Gaussian (LG) beams a promising candidate for high-dimensional quantum information [10], quantum memories [11], and quantum cryptography [12].

Atomic processes with twisted light beams, such as photoionization of atoms [13–15] and the scattering of twisted light beams by ions [16] or electrons [17] have also gained much attention. In addition to photoionization and scattering processes, the photoexcitation of atoms [18–20] by twisted light beams has attracted much interest in recent years. In a recent experiment, Schmiegelow *et al.* [21] observed for the first time the transfer of OAM from LG beams to the bound electrons of an atom during the excitation process. This experiment investigated the electric-quadrupole transition in the Ca<sup>+</sup> ion positioned on the beam axis and demonstrated the suppression of ac-Stark shift in the dark center of the LG beams. Later, a theoretical study [22] showed that the OAM is imprinted in the magnetic sublevel population and fluorescence following the excitation by a twisted light beam.

In contrast to the plane waves, twisted light beams support many different states of polarization, such as cylindrical polarization. However, photoexcitation of atoms by cylindrically polarized LG beams remains less explored. In this work, we analyze the multipole distribution of the cylindrically polarized LG beams and thus discuss its influence on the magnetic sublevel population in the target atom. Furthermore, for a target with Gaussian spatial distribution we calculate the total rate of excitation. Our calculation shows that the cylindrical polarization is more efficient than circular polarization in driving the electric-quadrupole transition in the target atoms. As an example, we consider the electric-quadrupole transition  $4s {}^{2}S_{1/2} \rightarrow 3d {}^{2}D_{5/2}$  in the Ca<sup>+</sup> ion. This particular transition has already been observed in the past experimental, as well as studied in theoretical works and thus serves as an ideal case for a comparison of our theoretical work.

This paper is structured as follows. In Sec. II A we first recall circularly polarized LG beams. We then use the knowledge of circularly polarized LG beams to understand the cylindrical polarization and to construct the vector potential of cylindrically polarized LG beams in Sec. II B. Moreover, we expand the vector potential of a LG beam into its multipole components to obtain complex weight factors for a given polarization in Sec. II C. In Sec. II D we use the multipole expansion of vector potential to derive the transition amplitude for the photoexcitation of atoms by cylindrically polarized LG beams. We evaluate the strength of the electric-quadrupole field in the beam cross section of a radially polarized LG beam with respect to a circularly polarized LG beam in Sec. III A. Furthermore, in Sec. III B we analyze the distribution of the projection of an electric-quadrupole field in the beam cross section with respect to the beam waist  $w_o$  and radial distance *b* from the beam axis of cylindrically polarized LG beams. In the first part of Sec. III C we illustrate the photoexcitation of a well-localized target by cylindrically polarized LG beams and in the second part we calculate the total rate of excitation for the electric-quadrupole transition between the  $4s {}^{2}S_{1/2} \rightarrow 3d {}^{2}D_{5/2}$  state in the Ca<sup>+</sup> ion target with a Gaussian spatial distribution. Finally, a summary of the paper is given in Sec. IV.

### **II. THEORETICAL BACKGROUND**

#### A. Circularly polarized LG beams

LG beams are paraxial twisted light beams whose amplitude distribution  $u(\mathbf{r})$  is known to satisfy the paraxial wave equation [23]

$$\nabla^2 u + 2ik\frac{\partial}{\partial z}u = 0. \tag{1}$$

This *paraxial wave* approximation is valid, if the amplitude distribution  $u(\mathbf{r})$  changes slowly with the distance z and this z dependence is less compared to variations of  $u(\mathbf{r})$  in the transverse direction:

$$\left|\frac{\partial^2 u}{\partial z^2}\right| \ll \left|2k\frac{\partial u}{\partial z}\right|, \quad \left|\frac{\partial^2 u}{\partial z^2}\right| \ll \left|\partial_t^2 u\right|. \tag{2}$$

In cylindrical coordinates the amplitude distribution of the LG beam is given by

$$u(\mathbf{r}) = \frac{1}{w(z)} \left( \frac{\sqrt{2}r}{w(z)} \right)^{m_{\ell}} \exp\left(-\frac{r^2}{w^2(z)}\right) L_p^{m_{\ell}} \left(\frac{2r^2}{w^2(z)}\right)$$
$$\times \exp\left[im_{\ell}\phi + \frac{ikr^2z}{2(z^2 + z_R^2)} - i(2p + m_{\ell} + 1)\arctan\left(\frac{z}{z_R}\right)\right], \tag{3}$$

where  $m_{\ell}$  is the projection of the OAM upon the propagation axis, p is the radial index of the LG beam,  $z_R$  is the (so-called) Rayleigh range, and w(z) is the beam width of the LG beam. The beam width w(z) of the LG beam varies along the propagation distance and is minimum for z = 0. This minimum beam width of a LG beam is known also as beam waist  $w_o$  $\equiv w(z = 0)$ . Moreover, Eq. (3) can be used to obtain the intensity distribution  $|u(\mathbf{r})|^2$  of the LG beam which exhibits a concentric ringlike structure in the beam cross section.

The wave amplitude of the circularly polarized LG beam in momentum space is expressed as a Fourier transformation of the amplitude distribution (3). Then the vector potential of a circularly polarized LG beam in Coulomb gauge is given by (see [22] for a detailed derivation)

$$\mathbf{A}_{m_{\ell},\lambda,p}^{(\text{circ})}(\mathbf{r}) = \int d^2 \mathbf{k}_{\perp} v_{pm_{\ell}}(k_{\perp}) e^{i(m_{\ell}+\lambda)\phi_{k}} \mathbf{e}_{k,\lambda} e^{i\mathbf{k}\cdot\mathbf{r}}, \quad (4)$$

where  $v_{pm_{\ell}}(k_{\perp})e^{i(m_{\ell}+\lambda)\phi_{k}}$  is the momentum space wave function and  $\mathbf{e}_{k,\lambda}e^{i\mathbf{k}\cdot\mathbf{r}}$  is the vector potential of a circularly polarized plane wave. In the above equation momentum space wave function  $v_{pm_{\ell}}(k_{\perp})$  is given by

$$v_{pm_{\ell}}(k_{\perp}) = \frac{(-i)^{m_{\ell}}}{w_{o}4\pi} e^{-k_{\perp}^{2}w_{o}^{2}/4} \left(\frac{k_{\perp}w_{o}}{2}\right)^{m_{\ell}} \sum_{\beta=0}^{p} (-1)^{\beta} 2^{\beta+m_{\ell}/2} \\ \times {\binom{p+m_{\ell}}{p-\beta}} L_{\beta}^{m_{\ell}} \left(\frac{k_{\perp}^{2}w_{o}^{2}}{4}\right).$$
(5)

As seen from Eq. (4), the LG beams with circular polarization can be expressed as a coherent superposition of circularly polarized plane waves in the momentum space. The momentum vector  $\mathbf{k}$  of these plane waves lies on the surface of a cone in the momentum space with an opening angle of  $\theta_k = \arctan(k_\perp/k_z)$ .

#### B. Cylindrically polarized LG beams

Cylindrically polarized LG beams can be constructed as a linear combination of two circularly polarized LG modes [24]. This results in nonseparable spatial and polarization modes [25]. As a consequence of nonseparable spatial and polarization modes, the state of polarization across the beam cross section in cylindrically polarized LG beams is spatially inhomogeneous [26]. Beams which are linear combinations of two LG modes are known as vector beams [27] and constitute a vector solution to the paraxial wave equation.

Radial and azimuthal polarizations are two special cases of a cylindrical polarization constructed as a linear combination of two LG modes with the projection of OAM  $m_{\ell} = \pm 1$ and helicity  $\lambda = \mp 1$  [28]. The electric field of the radially polarized LG beam always points in the radial direction and is perpendicular to the beam axis [29,30]. The vector potential of a radially polarized LG beam is constructed as a linear combination of vector potential of right circularly polarized  $\mathbf{A}_{m_{\ell}=-1,\lambda=-1,p}^{(circ)}(\mathbf{r})$  and left circularly polarized  $\mathbf{A}_{m_{\ell}=1,\lambda=-1,p}^{(circ)}(\mathbf{r})$ LG beams and is given by

$$\mathbf{A}_{p}^{(\mathrm{rad})}(\mathbf{r}) = \frac{-i}{\sqrt{2}} \Big[ \mathbf{A}_{m_{\ell}=1,\lambda=-1,p}^{(\mathrm{circ})}(\mathbf{r}) + \mathbf{A}_{m_{\ell}=-1,\lambda=1,p}^{(\mathrm{circ})}(\mathbf{r}) \Big].$$
(6)

Similarly, the electric field direction of the azimuthally polarized LG beams is always perpendicular to the radial direction [30,31] and its vector potential is given by

$$\mathbf{A}_{p}^{(\text{azmth})}(\mathbf{r}) = \frac{1}{\sqrt{2}} \Big[ \mathbf{A}_{m_{\ell}=1,\lambda=-1,p}^{(\text{circ})}(\mathbf{r}) - \mathbf{A}_{m_{\ell}=-1,\lambda=1,p}^{(\text{circ})}(\mathbf{r}) \Big].$$
(7)

Further, we use the vector potential of the LG beam of a given polarization to obtain the complex weight factors using the multipole expansion in the next section.

#### C. Multipole expansion of a vector potential of the LG beam

A multipole expansion of the radiation field enables us generally to expand the vector potential in angular momentum basis [32,33]. Since the intensity distribution of the LG beam is nonuniform in the beam cross section [34], we perform multipole expansion of the vector potential of a LG beam at a radial distance *b* from the beam axis. That is, when the *z* axis is translated by a vector  $\mathbf{b} = b\mathbf{e}_x$  from the beam axis. Then the multipole expansion of such a vector potential at a position *b* from the beam axis, for a circularly polarized LG beam is

given by

$$\mathbf{A}_{m_{\ell},\lambda,p}^{(\text{circ})}(\mathbf{r};b,w_o) = \sum_{L,M,\Lambda} W_{m_{\ell},\lambda,p}^{(\text{circ})}(L,M,\Lambda;b,w_o) \, \mathbf{a}_{L,M}^{\Lambda}(\mathbf{r}),$$
(8)

where L and M are the eigenvalues of the total angular momentum (TAM) and the projection of TAM operators, respectively. The  $W_{m_{\ell}\lambda,p}^{(circ)}(L, M, \Lambda; b, w_o)$  is the complex weight factor of the expansion which depends on the radial distance from the beam axis *b*, the beam waist  $w_o$ , projection of orbital angular momentum  $m_\ell$ , the radial index *p*, and the helicity  $\lambda$ . The multipole expansion expresses the vector potential of the LG beam as a linear combination of electric ( $\Lambda = 1$ ) and magnetic ( $\Lambda = 0$ ) multipole components  $\mathbf{a}_{L,M}^{\Lambda}(\mathbf{r})$ . Mathematically,  $\mathbf{a}_{L,M}^{\Lambda}(\mathbf{r})$  are expressed in terms of vector spherical harmonics of rank *L* [35,36]. For circularly polarized LG beams, the complex weight factors are given by

$$W_{m_{\ell},\lambda,p}^{(\text{circ})}(L,M,\Lambda;b,w_{o}) = \sum_{L,M,\Lambda} \sum_{\beta=0}^{p} (i\lambda)^{\Lambda} (-1)^{\beta} 2^{\beta+(m_{\ell}/2)} {p+m_{\ell} \choose p-\beta} \frac{(-i)^{m_{\ell}} w_{o}}{2\pi} (i)^{L+m_{\ell}+\lambda-M} (2L+1)^{1/2} \\ \times \int_{0}^{\infty} k_{\perp} dk_{\perp} d_{M,\lambda}^{L}(\theta_{k}) e^{-k_{\perp}^{2} w_{o}^{2}/4} {\left(\frac{k_{\perp} w_{o}}{2}\right)}^{m_{\ell}} L_{\beta}^{m_{\ell}} \left(\frac{(k_{\perp} w_{o})^{2}}{4}\right) J_{m_{\ell}+\lambda-M}(k_{\perp}b).$$
(9)

Using these, we define the complex weight factors for the radially polarized LG beam as

$$W_{p}^{(\text{rad})}(L, M, \Lambda; b, w_{o}) = \frac{-i}{\sqrt{2}} \Big[ W_{m_{\ell}=1,\lambda=-1,p}^{(\text{circ})}(L, M, \Lambda; b, w_{o}) + W_{m_{\ell}=-1,\lambda=1,p}^{(\text{circ})}(L, M, \Lambda; b, w_{o}) \Big],$$
(10)

and for the azimuthally polarized LG beam as

$$W_{p}^{(\text{azmth})}(L, M, \Lambda; b, w_{o}) = \frac{1}{\sqrt{2}} \Big[ W_{m_{\ell}=1,\lambda=-1,p}^{(\text{circ})}(L, M, \Lambda; b, w_{o}) - W_{m_{\ell}=-1,\lambda=1,p}^{(\text{circ})}(L, M, \Lambda; b, w_{o}) \Big].$$
(11)

With the help of complex weight factors, we can study the strength of the individual multipole components of the radiation field. Since these complex weight factors depend on the radial distance *b* and the beam waist  $w_o$ , we can control the multipole distribution of the LG beam by carefully choosing *b* and  $w_o$ .

#### D. Interaction of LG beams with the target atom

The transition between the initial  $|\alpha_i J_i M_i\rangle$  and the final  $|\alpha_f J_f M_f\rangle$  bound states of an atom is given by the transition amplitude  $M_{fi}$ 

$$M_{fi} = \langle \alpha_f J_f M_f | \boldsymbol{\alpha} \cdot \mathbf{A}(\mathbf{r}) | \alpha_i J_i M_i \rangle, \qquad (12)$$

where  $\mathbf{A}(r)$  is the vector potential of either radially polarized or azimuthally polarized LG beams,  $\boldsymbol{\alpha}$  is the Dirac matrix, and the atomic initial (final) states are characterized by TAM  $J_i$  $(J_f)$ , TAM projection  $M_i(M_f)$  quantum numbers, and  $\alpha$  refers to all additional quantum numbers, respectively.

We substitute the multipole expansion of the vector potential of LG beams into Eq. (12) to obtain the transition amplitude

$$M_{fi}(b, w_o) = \sum_{L,M,\Lambda} W(L, M, \Lambda; b, w_o) \\ \times \langle \alpha_f J_f M_f | \boldsymbol{\alpha} \cdot \mathbf{a}_{L,M}^{\Lambda}(\mathbf{r}) | \alpha_i J_i M_i \rangle, \qquad (13)$$

where  $W(L, M, \Lambda; b, w_o)$  is the complex weight factor of either a radially polarized or azimuthally polarized LG beam. The rest of the transition amplitude equation is solved using the Wigner-Eckart theorem [32,33], which gives the transition amplitude

$$M_{fi}(b, w_o) = \sum_{L,M,\Lambda} W(L, M, \Lambda; b, w_o) \langle J_i M_i, LM | J_f M_f \rangle$$
$$\times \langle \alpha_f J_f || \boldsymbol{\alpha} \cdot \mathbf{a}_L^{\Lambda}(\mathbf{r}) || \alpha_i J_i \rangle$$
(14)

as the product of geometrical and atomic factors. The geometrical properties are described by Clebsch-Gordan (CG) coefficients and the complex weight factors  $W(L, M, \Lambda; b, w_o)$ . Similarly, the reduced matrix elements describe the atomic properties. The atomic excitation process is influenced by both geometrical and atomic properties. It is clear from the above Eq. (14) that the transition amplitude depends on the complex weight factors  $W(L, M, \Lambda; b, w_o)$ , which together with the CG coefficients and reduced matrix elements determine the amplitudes of the individual transitions.

The transition amplitude of the atomic transition between two bound states must satisfy the following set of rules, known as *selection rules*, given by

$$M_i + M = M_f, \tag{15}$$

$$|J_f - J_i| \leqslant L \leqslant |J_f + J_i|, \tag{16}$$

$$\pi_i \pi_f = (-1)^{L+p+1}.$$
(17)

Here  $\pi_i \pi_f$  are the parity of the initial and final atomic states.

## **III. RESULTS AND DISCUSSION**

#### A. Distribution of electric-quadrupole field in the beam cross section of cylindrically polarized LG beams

We now use the complex weight factors  $W_{m_{\ell},\lambda,p}(L, M, \Lambda; b, w_o)$  to analyze the distribution of the electric-quadrupole field in the beam cross section of cylindrically polarized LG beams. The strength of a multipole component *L*, in particular the electric-quadrupole



FIG. 1. The relative strength of the electric-quadrupole field  $S_r(E2)$  is plotted against the radial distance b from the beam axis of the LG beam. The top plot shows variation of  $S_r(E2)$  for three different radial indices p = 0, 1, 2. The plot describes the variation of the strength of the electric-quadrupole field of radial polarization with respect to circular polarization in the beam cross section of the LG beam with beam waist of  $w_o = 2.7 \ \mu m$ . The bottom plot describes the intensity profile of the LG beams with radial index p = 0, 1, 2.

component (L = 2), in the beam cross section is given  $\sum_{M=-L}^{+L} |W_{m_{\ell},\lambda,p}(L=2, M, \Lambda=1; b, w_o)|^2$ for by а corresponding polarization of the LG beam. We define the relative strength of the electric-quadrupole field

$$S_r(E2) = \frac{\sum_{M=-L}^{+L} |W_{m_\ell,\lambda,p}^{(\text{circ})}(L=2, M, \Lambda=1; b, w_o)|^2}{\sum_{M=-L}^{+L} |W_p^{(\text{rad})}(L=2, M, \Lambda=1; b, w_o)|^2}$$
(18)

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as a ratio of strength of the electric-quadrupole field between circularly polarized and radially polarized LG beams.

In Fig. 1, we plot  $S_r(E2)$  against the radial distance b from the beam axis of a LG beam for different radial index p values in the top panel and the corresponding intensity profile of LG beams in the bottom panel. For the radial index p = 0, we observe the value of  $S_r(E2)$  to be greater than 1 near the beam axis  $(b \approx 0)$  and decreases rapidly to less than 1 as radial distance b increases. The behavior of the ratio  $S_r(E2)$ indicates that the strength of the electric-quadrupole field is suppressed near the beam axis of the cylindrically polarized LG beam. However, for large b values the ratio  $S_r(E2)$  is less than 1, indicating a strong electric-quadrupole field in the beam cross section of the cylindrically polarized LG beam.

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The radial index p of the LG beam modifies the strength of the electric-quadrupole field in the beam cross section as described by Fig. 1 for p = 1, 2. Similar to the p = 0 case, the ratio  $S_r(E2)$  is greater than 1 near the beam axis, indicating the suppression of an electric-quadrupole field near the beam axis for the cylindrically polarized LG beam. For large b values, we observe the ratio  $S_r(E2)$  to be less than 1 near the dark region in the beam cross section of the LG beam. The vertical lines in Fig. 1 are used to denote the corresponding dark region in the beam cross section of the LG beam with the help of an intensity profile. In contrast to the p = 0 case, cylindrically polarized LG beams possess a dominant electric-quadrupole field with respect to circularly polarized LG beams only near the dark region in the off-axis region.

The electric-quadrupole component can be associated with the electric field gradient of the light beam and is responsible for driving the electric-quadrupole transition in the target atoms. Therefore, the region in Fig. 1 with a value of  $S_r(E2)$ less than 1 describes a strong electric field gradient region in the beam cross section of cylindrically polarized LG beams. This suggests that, if we were to place an atom in such a region the electric-quadrupole transition would be more efficiently driven by a cylindrical polarization over a circularly polarized LG beam.

## B. Influence of the beam waist and radial position on the projection of TAM of LG beams

We shall now discuss the strength of the projection of multipole component M in the beam cross section of the LG beam using the complex weight factors. As seen from the complex weight factors, the multipole distribution of the LG beam varies with the beam waist  $w_{a}$  and the radial distance b from the beam axis. To understand the variation of the projection of TAM across the beam cross section, we define the relative weight of projection of the electric-quadrupole field as

$$W_r(M) = \frac{|W_p(L=2, M, \Lambda=1; b, w_o)|^2}{\sum_{M=-L}^{+L} |W_p(L=2, M, \Lambda=1; b, w_o)|^2},$$
 (19)

where  $|W_p(L=2, M, \Lambda = 1; b, w_o)|^2$  denotes the modulus squared of the complex weight factor of a radially or azimuthally polarized LG beam.

In Fig. 2, we plot  $W_r(M)$  against the radial distance from the beam axis b while keeping the beam waist fixed at 2.7  $\mu$ m [21]. For radially polarized LG beams, on the beam axis we observe the M = -1 component to be dominant while the M = 0 component is dominant for the azimuthally polarized LG beam. For large values of b, we notice the M = +1component to be dominant for both radially and azimuthally polarized LG beams.

Similarly in Fig. 3, we plot  $W_r(M)$  against the beam waist  $w_o$  of the cylindrically polarized LG beam for a fixed b value. Since we consider a paraxial LG beam, the minimum beam waist is given by the diffraction limit

$$w_o^{(\min)} = \frac{\lambda}{\sqrt{2}\pi},\tag{20}$$

where  $\lambda = 729$  nm corresponds to the wavelength of the LG beam used for the driving transition between the 4s and 3dstates of a Ca<sup>+</sup> ion [21]. For this beam waist  $w_o = 0.16 \ \mu \text{m}$ ,



FIG. 2. The relative weight of the projection of the quadrupole component  $W_r(M)$  is plotted against the radial distance *b* from the beam axis for the radially polarized (top) and the azimuthally polarized LG beam (bottom). In the above plots radial index p = 0 and the beam waist is fixed to  $w_o = 2.7 \ \mu \text{m}$ .

we observe the M = +1 component to be dominant in both radial and azimuthal polarization. For larger beam waist  $w_o$ , the M = -1 and M = 0 components dominate in radially and azimuthally polarized LG beams, respectively.

Thus, by carefully selecting the beam waist  $w_o$  and radial distance *b* we can control the relative strength of *M* in the beam cross section of the LG beam of either radial or azimuthal polarization.

## C. Photoexcitation of Ca<sup>+</sup> ion by LG beams

# 1. Single ion target

We now illustrate the photoexcitation of a well-localized target by a cylindrically polarized Laguerre-Gaussian beam of beam waist  $w_o$  and radial index p with the help of Figs. 2 and 3 and the selection rule  $M_i + M = M_f$ . For example, we consider the electric-quadrupole transition between the  $|4S_{1/2}, M_i = -1/2\rangle$  and  $|3D_{5/2}, M_f = \pm 5/2\rangle$  states in a single Ca<sup>+</sup> ion driven by cylindrically polarized LG beams of wavelength 729 nm [21].

For a single Ca<sup>+</sup> ion positioned along the beam axis, a radially polarized LG beam drives the transition between  $|4S_{1/2}, M_i = -1/2\rangle$  and the  $|3D_{5/2}, M_f = -3/2\rangle$  magnetic substate transferring the M = -1 component, as shown in



FIG. 3. The relative weight of the projection of quadrupole component  $W_r(M)$  is plotted against the beam waist  $w_o$  for the radially polarized (top) and the azimuthally polarized LG beam (bottom). In the above plots radial index p = 0 and the radial distance is fixed to  $b = 0.5 \ \mu m$ .

Fig. 2. As the radial distance *b* between the target Ca<sup>+</sup> ion and the beam axis is increased, we observe a transition between  $|4S_{1/2}, M_i = -1/2\rangle$  and the  $|3D_{5/2}, M_f = 1/2\rangle$  magnetic substate absorbing M = +1 component. For the target Ca<sup>+</sup> ion positioned in the off-axis region, say,  $b = 0.5 \ \mu\text{m}$  interacting with a radially polarized LG beam of minimum beam waist  $w_o = 0.16 \ \mu\text{m}$ , we observe a transition between  $|4S_{1/2}, M_i = -1/2\rangle$  and the  $|3D_{5/2}, M_f = +1/2\rangle$  magnetic substate absorbing M = +1 component as shown in Fig. 3. But as we increase the beam waist  $w_o$ , the strength of the M = +1component in the beam cross section of the radially polarized LG beam decreases rapidly and the strength of the M = -1component increases. This results in the transition between the  $|4S_{1/2}, M_i = -1/2\rangle$  and  $|3D_{5/2}, M_f = -3/2\rangle$  magnetic substates.

Similar to the above discussion, we can explain the photoexcitation of a well-localized Ca<sup>+</sup> ion and the corresponding atomic transitions driven by the azimuthally polarized LG beam.

#### 2. Macroscopic target

The well-localized pointlike target considered in Sec. III C 1 is an idealistic model and is difficult to realize in the experimental setups. In experiments, even a single ion target trapped in Paul traps has a thermal spatial spread [21]. Hence, we now consider the atomic target to have a Gaussian



FIG. 4. Logarithmic plot of the total rate of excitation for the electric-quadrupole transition between the  $4s \, {}^{2}S_{1/2} \rightarrow 3d \, {}^{2}D_{5/2}$  levels of the Ca<sup>+</sup> ion driven by a LG beam is plotted as a function of the width of the target  $\sigma$  for azimuthal (green solid line), radial (brown dashed line), and circular polarization (blue dotted line). In the top figure the target is placed on the beam axis  $b_{o,x} = 0$  and in the bottom figure the target is displaced from the beam axis by  $b_{o,x} = 2 \, \mu$ m. In both the plots, radial index p and beam waist  $w_o$  are kept fixed at 0 and 2.7  $\mu$ m, respectively.

spatial distribution as observed in the experiments [21,37] and which is given by

$$f(\boldsymbol{b}) = \frac{1}{2\pi\sigma^2} \exp\left(-\frac{(\boldsymbol{b} - \boldsymbol{b}_o)^2}{2\sigma^2}\right), \quad (21)$$

where  $\sigma$  is the width of the target and vector  $\boldsymbol{b}_o = b_o \boldsymbol{e}_x \equiv b_{o,x}$ is the distance from the beam axis to the center of the target. For an atomic target with a Gaussian spatial distribution interacting with the Laguerre-Gaussian beams we define the total rate of excitation as

$$W_{fi}^{\text{LG}} = \frac{2\pi}{\alpha^2} \sum_{M_f} \int d^2 \boldsymbol{b} f(\boldsymbol{b}) |M_{fi}(b, w_o)|^2.$$
(22)

Since we consider a target prepared in a specific magnetic state  $M_i$ , we do not sum and average over all initial states. As seen from Eqs. (21) and (22) the rate is proportional to the modulus squared of the transition amplitude and the Gaussian distribution of the target. The full many-electron transition amplitude for the electric-quadrupole transition (4*s*  ${}^2S_{1/2} \rightarrow 3d {}^2D_{5/2}$ ) was calculated using the JAC [38] package (and GRASP [39]).

In Fig. 4 we plot the total rate of excitation for electricquadrupole transition as a function of the width of the target



FIG. 5. Logarithmic plot of the total rate of excitation for the electric-quadrupole transition between  $4s {}^{2}S_{1/2} \rightarrow 3d {}^{2}D_{5/2}$  levels of the Ca<sup>+</sup> ion driven by the LG beam is plotted as a function of the width of the target  $\sigma$  for azimuthal (green solid line), radial (brown dashed line), and circular (blue dotted line) polarization. In the top figure the target is placed on the beam axis  $b_{o,x} = 0$  and in the bottom figure the target is displaced from the beam axis by  $b_{o,x} = 2 \mu m$ . In both the plots, radial index p and beam waist  $w_{o}$  are kept fixed at 0 and 0.16  $\mu$ m, respectively.

for the beam waist of the cylindrically polarized LG beam of  $w_o = 2.7 \ \mu m$ . In addition, we show the total rate of excitation for circularly polarized LG beams as a comparison. The width of the target  $\sigma$  is varied from 0 to a maximum of 0.06  $\mu$ m as observed in the experiments. For the target placed on the beam axis, we observe the azimuthally polarized LG beam to be more efficient in driving the electric-quadrupole transition for a target with small as well as large width  $\sigma$ . In the bottom of Fig. 4, we consider the target to be placed near the maximum intensity region in the beam cross section, that is, when the x component of  $\boldsymbol{b}_o(b_{o,x}) = 2 \ \mu \text{m}$ . For such a case, we observe both circular and cylindrical polarization to have the same  $W_{fi}^{LG}$  for a small width of the target. But as we increase the width of the target, we observe  $W_{fi}^{LG}$  to decrease rapidly for both the cylindrical as well as the circularly polarized LG beam. However, for larger width of the target we notice  $W_{fi}^{LG}$  to be more for cylindrical polarization than for circular polarization. As shown in Fig. 1 the strength of the electric-quadrupole field for the cylindrically polarized LG beam increases as we move away from the beam axis, hence we obtain a maximum  $W_{fi}^{LG}$  than circular polarization.

In Fig. 5 the variation of the total rate of excitation is plotted as a function of the width of the target placed in the

center of the beam and displaced from the beam axis for the minimum possible beam waist  $w_o = 0.16 \ \mu\text{m}$ . Similar to the case for beam waist  $w_o = 2.7 \ \mu\text{m}$ , we observe the azimuthal polarization to have a maximum total rate of excitation when compared to circular and radial polarization as shown in the top figure. In contrast to the beam waist  $w_o = 2.7 \ \mu\text{m}$ , we observe a higher total rate of excitation for radial polarization when we consider  $w_o = 0.16 \ \mu\text{m}$ . For the target placed in the region of maximum intensity, we observe that both radial and azimuthal polarization yield the same total rate of excitation. Furthermore, we can observe that the  $W_{fi}^{\text{LG}}$  for cylindrical polarization when the target is placed in the maximum intensity region of the beam with beam waist  $w_o = 0.16 \ \mu\text{m}$ .

## **IV. SUMMARY**

We have theoretically investigated the photoexcitation of atoms by LG beams especially for cylindrical polarization. To do so, we constructed the complex weight factor of cylindrically polarized LG beams as a linear combination of complex weight factors for circular polarization. We analyzed the strength of the electric-quadrupole field across the beam cross section of cylindrically polarized LG beams. We observed that the strength of the electric-quadrupole field of cylindrical polarization is maximum away from the beam axis. In addition, we observed that the strength of the electric-quadrupole field in the beam cross section is sensitive to the radial index p of the LG beam. The variation of the magnetic component of the electric-quadrupole field was analyzed as a function of beam waist  $w_o$  and the radial distance b from the beam axis. To better understand the variation of the magnetic component of the electric-quadrupole field, we plotted the relative weight of the projection of electric-quadrupole  $W_r(M)$  against the beam waist and the radial distance from the beam axis.

Furthermore, we illustrated the effects of varying multipole distribution on the magnetic sublevel population in the target atom. As an example, we considered the electric-quadrupole transition in the target Ca<sup>+</sup> ion. In addition, we calculated total rate of excitation of electric-quadrupole transition  $(4s \ ^2S_{1/2} \rightarrow 3d \ ^2D_{5/2})$  in the target Ca<sup>+</sup> ion with a Gaussian spatial distribution. We observed that the total rate of excitation for the target Ca<sup>+</sup> ion excited by the cylindrically polarized LG beam varies with beam waist  $w_o$  and the distance between the center of the target and the beam axis  $b_{o,x}$ . Our calculation explicitly shows that the cylindrically polarized LG beam is more efficient than circular polarization for driving an electric-quadrupole transition in a mesoscopic target.

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