Interaction of a Laguerre–Gaussian beam with trapped Rydberg atoms

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Abstract

Previous studies show that within paraxial limit and electric dipole approximations, the orbital angular momentum (OAM) of a Laguerre–Gaussian (LG) beam rotates the whole atom about the beam axis but does not affect the internal electronic motion. The contribution of the Gaussian part of the LG beam profile to the angular momentum exchange is not usually taken into account. In this paper, we develop a theory which shows that not only the OAM of a LG beam, but also the geometry of the beam cross section, plays an important role in the electronic lowest-order transitions in the interaction of the beam with a trapped Rydberg atom, due to the large span of its electronic wavefunctions. In this interaction, the standard dipole selection rules do not hold and our results show that the otherwise forbidden transitions can become sufficiently probable.

Keywords: Laguerre–Gaussian beam, Rydberg atoms, selection rules

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

1. Introduction

We are living in an era where quantum technology is blooming and it is important to find suitable systems which can carry and exchange quantum information. Among them, photon–ion, photon–atom and photon–molecule systems are propitious. In a normal Hermite–Gaussian laser beam, each photon carries only a single unit of angular momentum in its polarization or spin property [1]. But this limitation can be circumvented by using Laguerre–Gaussian (LG) beams which, in addition to the spin angular momentum (SAM), carry quantized orbital angular momentum (OAM) associated with its azimuthal phase structure [2]. There is no upper limit on the value of OAM and an enormous amount of quantum data can be encoded in it. The OAM of light is also termed the topological charge (TC) of the optical vortex or helical phase front, which can be created by dislocations in a diffractive structure. Due to its unbounded quantum numbers representing the states of OAM, the LG beam has found a great number of uses in the fundamental studies of quantum systems [3–8], optical communications [9–11], the detection of spinning objects [12], the generation of second and higher harmonics [13–17], wave-mixing [18], the generation of singular optical lattices [19] etc. An LG beam is used to generate optical tweezers for trapping and rotating micron-sized particles [20–23].

Optical OAM is used to create and manipulate vortex states of atomic Bose–Einstein condensate (BEC) [24–31]. Recent works suggest that under paraxial approximation, optical OAM can influence internal electronic transitions of ultracold atoms only beyond the dipole transition [31]. The reason for this is that the atomic dimension is far too small for internal electronic motion to feel the phase distribution over the beam cross section. However, the intensity gradient over the beam cross section affects the quadrupole transition [32, 33]. But quadrupole transition rates are orders of magnitude weaker than the dipole rates and hence it is difficult to observe the quadrupole effect.

In this work, we theoretically study the interaction of optical OAM with a trapped Rydberg atomic system whose dimension is large enough to feel the phase distribution of the LG beam. Rydberg atoms are currently of tremendous interest...
because of their strong dipole–dipole interactions and long lifetimes [34, 35] leading to applications in dipole blockades [36], quantum gates [37], the realization of few-photon nonlinearities [38, 39], the single photon switch [40], electromagnetically induced transparency [41], electromagnetically induced absorption [42], performing precision measurements [43, 44], electrometry [45, 46] etc. Furthermore, Rydberg atom trapping has been investigated both theoretically [47] and experimentally [48]. A recent study [49] shows the violation of the standard selection rules of the electromagnetic transitions in the interaction of thermal Rydberg atoms with the LG beam. Here we consider a Rydberg atom with a typical size of 0.2 μm, trapped in a harmonic potential whose size is one order of magnitude larger than the atomic size. The cross section of the LG beam is assumed to be comparable with the trap size. Hence, not only the phase factor but also the Gaussian portion of the beam cross section influences the electronic transitions. The contribution of the Gaussian portion is generally neglected because the size of a normal atom is too small to feel the Gaussian factor of the beam, in the case of the atom being placed near the beam axis. We show that angular momentum coming from the topological charge (TC) allows us access to the otherwise forbidden transitions (2Σ_{1/2} → 2 D_{5/2}).

This paper is organized in the following way. In section 2, we develop a theory to show how the angular momenta from different factors of the LG beam are transferred in the interaction with a trapped Rydberg atom in two dimensional harmonic potential. We describe only the first order effect of the Gaussian factor for simplicity and relevance. We calculate the transition matrix elements between electronic states. We numerically calculate and analyze the transition Rabi frequencies in section 3. We study the variation of the strengths of these transitions (in terms of the Rabi frequencies) with respect to the TC of the beam. Finally, we conclude in section 4 with remarks on the prospect of this work.

2. Theory

For LG mode, \( \mathbf{E}(r, t) = \mathcal{E}(r) e^{i \omega t} \hat{c} \). The field amplitude in spherical coordinates is expressed as

\[
\mathcal{E}(r) = \mathcal{E}_0 \frac{1}{w_0^l} \frac{2^{|l|+1}}{\pi|l|!} (r \sin \theta)^{|l|} e^{i \theta l} e^{-\frac{2 r^2}{w_0^2}} e^{i \varphi r} \cos \theta, \tag{1}
\]

where \( \omega \) is the frequency and \( \hat{c} \) is the unit polarization vector of the beam with \( l \) as the OAM [50]. Here \( w_0 \) is the radius of the beam and the parameter \( \mathcal{E}_0 = \sqrt{2 l + 1} \) with \( l \), \( c \), \( \phi_0 \) being the intensity, velocity of light and electric permittivity of free space, respectively. The beam axis is taken as the \( z \)-axis of the laboratory coordinate system. In terms of solid spherical harmonics \( \mathcal{R}_{m_l}^{|l|} \) defined in the appendix, equation (1), can be expressed as

\[
\mathcal{E}(r) = \mathcal{E}_0 \sum_{q=0}^{\infty} \frac{2^{|l|+1}}{\pi|l|!} (r \sin \theta)^{|l|} e^{i \theta l} e^{-\frac{2 r^2}{w_0^2}} e^{i \varphi r} \cos \theta, \tag{2}
\]

Equation (2) shows two variables; one is TC \( l \) and the other one is \( q \), coming from the Gaussian factor of the field distribution. It is the order of the summation series of the exponential profile of the beam. The appearance of both \( q \) and \( -q \) confirms that no net angular momentum is imparted by the Gaussian term. Using the relations of solid harmonics discussed in the appendix, we obtain

\[
f(l, q) = \frac{4 q !}{w_0^2 r^{|l|+1} (2 q !) ! 2 |l| !} \sqrt{(2 ^{|l|+1} |l| !) / \pi}. \]

The interaction Hamiltonian is given in the Power–Zienau–Wooley (PZW) scheme [29, 51] as

\[
H_{int} = - \int \mathcal{P}(r) \mathcal{E}(r', t) dr'. \tag{3}
\]

The polarization vector, \( \mathcal{P}(r) \), in closed integral form, is defined by

\[
\mathcal{P}(r') = - \varepsilon r \int_0^l \delta(r' - r_{CM} - \lambda m_c / m_l) d \lambda. \tag{4}
\]

The relative coordinate (internal coordinate) of the electron in the CM frame of the atom is \( r = r_c - r_{CM} \), where \( r_c \) and \( r_{CM} \) are the electronic and CM coordinates, respectively. \( m_c \) and \( m_l \) are masses of the atomic core and the whole atom, respectively. Here, \( m_c \) and \( m_l \) are considered to be the same due to the smallness of the mass of the electron.

Substituting equation (2) and equation (4) into equation (3), \( H_{int} \) takes the form

\[
H_{int} = \varepsilon r \int_0^l \mathcal{E}(r_{CM} + \lambda r) e^{i \varphi r} d \lambda \tag{5}
\]

\[
= \varepsilon \mathcal{E}_0 \int_0^l \sum_{q=0}^{\infty} f(l, q) \mathcal{A}(l, q) A(l, -q) e^{i l \varphi r} e^{i \varphi r} d \lambda \tag{6}
\]

where

\[
\mathcal{A}(l, \lambda, r, r_{CM}) = \sum_{l=0}^l \sum_{m_{CM}} R_{m_C}^{|l|} (\lambda r) \mathcal{R}_{m_{CM}}^{|l|} (r_{CM}). \tag{7}
\]

The angular factor \( e^{i l \varphi r} \) here depends only on the variation in electric field along the direction of propagation, which is negligible in the paraxial limit. In this work, we are rather interested in studying the variation of field over the cross section. Hence, we take, \( e^{i l \varphi r} \) is substituted by \( r \sqrt{\frac{2 \lambda}{2 \pi \sum_{\sigma=0,1,-1} \epsilon_\sigma Y_{\lambda}(\theta_c, \phi_0)} \), where \( \epsilon_\sigma = (E_e \pm i E_e)^{1/2} \) and \( \epsilon_0 = E_e. \) After the integration over \( \lambda \), the interaction Hamiltonian takes the form:

\[
H_{int} = \varepsilon \mathcal{E}_0 \sum_{l=0}^l \sum_{m_{CM}} \sum_{\sigma=0,1,-1} \epsilon_\sigma \mathcal{F}_e \mathcal{F}_{CM} \tag{8}
\]

where \( \mathcal{F}_e \) and \( \mathcal{F}_{CM} \) are the angular components of the interaction
Hamiltonian containing spherical harmonics involving the electron and the CM angular coordinates, respectively. \( J_2(q; b, c, z) \) is a hypergeometric function. We should mention here that the angular momentum operators \( l_i \) and \( l_j \) correspond to the TC of the beam while \( q_i, l_2, l_3 \) depend on the Gaussian factor of the beam cross section. \( m, m_1, m_2, m_3 \) are respective projections of \( l_i, l_2, l_3 \) along the z-axis, \( \sigma \) denotes angular momentum associated with polarization of the beam. It is evident that the TC and the Gaussian term contribute to both the CM and electronic angular momentum but the polarization contributes only to the electronic motion. Two important parameters of the radial matrix elements of the electronic part and the CM part are \( \alpha = l_i + l_2 + l_3 + 1 \) and \( \beta = |l| + 2q - l_i - l_2 - l_3 \), respectively. The expression for \( \alpha \) shows that if any one of \( l_i, l_2, l_3 \) has a non-zero value, which is obvious in the present case, then the standard dipole approximation is broken. We write the transition matrix element between the initial and final composite (electronic plus CM) states as

\[
\mathcal{M}_{i\rightarrow f} = \langle \tilde{\Psi}|H_{\text{int}}|\tilde{\Psi} \rangle = eE_0 \left[ \sum_{\alpha=0,1} \sum_{q=0}^{\infty} \sum_{l_i=0}^{l_i} \sum_{l_2=0}^{l_3} \sum_{l_3=0}^{l_3} \epsilon_{\alpha} (l, q) \Gamma \left( \frac{\alpha}{2} \right) \right.
\]

\[
\times C_{l_i,q,l_2,l_3}^{m_i,m_2,m_3} \langle \psi_{\text{CM}}^l (r_{\text{CM}}) \rangle \left| \psi_i \right\rangle 
\]

\[
\times \langle \tilde{\Psi} | \frac{r}{w_r} \rangle \alpha \left( \frac{w_r}{w_0} \right)^{2q+1} \left( \frac{2q!}{(2q+1)!} \right) \left( \frac{2^3 (l+1)!}{3} \right) \times \delta_{M_i,l-m_i-m_2-m_3+M_f}
\]

\[
\times \delta_{M_i,l-m_i-m_2-m_3+M_f} \times \delta_{M_i,l-m_i-m_2-m_3+M_f}
\]

\[
= \left[ \frac{\hbar^2}{2\mu} \frac{d^2}{d^2r} + \frac{2d}{r \, dr} + \frac{\hbar^2}{2\mu r^2} + V(r) \right] \phi_e(r)
\]

Let us now analyze \( \mathcal{M}_{i\rightarrow f} \) at the lowest order transition level.

\[
\mathcal{M}_{i\rightarrow f} = \langle \tilde{\Psi}|H_{\text{int}}|\tilde{\Psi} \rangle = eE_0 \sum_{\alpha=0,1} \sum_{q=0}^{\infty} \sum_{l_i=0}^{l_i} \sum_{l_2=0}^{l_3} \sum_{l_3=0}^{l_3} \epsilon_{\alpha} (l, q) \Gamma \left( \frac{\alpha}{2} \right)
\]

\[
\times C_{l_i,q,l_2,l_3}^{m_i,m_2,m_3} \langle \psi_{\text{CM}}^l (r_{\text{CM}}) \rangle \left| \psi_i \right\rangle 
\]

\[
\times \left( \frac{r}{w_r} \right)^{\alpha-1} \left| \psi_i \right\rangle
\]

\[
\times \langle \tilde{\Psi} | \frac{r}{w_r} \rangle \alpha \left( \frac{w_r}{w_0} \right)^{2q+1} \left( \frac{2q!}{(2q+1)!} \right) \left( \frac{2^3 (l+1)!}{3} \right) \times \delta_{M_i,l-m_i-m_2-m_3+M_f}
\]

Equation (10) shows that the sign of the TC of the beam (l) has an effect on the angular portion of the CM or electronic transition matrix element, but not on the radial parts as expected. In the next section, we numerically calculate the Rabi frequencies of possible transitions defined as \( \Omega_{i\rightarrow f} = \mathcal{M}_{i\rightarrow f}/\hbar \).

3. Results and discussions

The total wavefunction of the Rydberg atom has two parts, the CM wavefunction \( \psi_{\text{CM}}(r_{\text{CM}}) \) and the electronic wavefunction \( \psi_e(r) \). The CM wavefunction depends on the external trapping potential and in a two dimensional harmonic oscillator potential it takes the form

\[
\psi_{\text{CM}}(r_{\text{CM}}, \phi) = A_{N,M}(r_{\text{CM}}) e^{iM\phi},
\]

Here the normalized amplitude \( A_{N,M}(x) = \frac{1}{w_r} \frac{2n^2}{\pi^{1/4}} \times L_\nu^M (x^2) e^{-x^2/2} \) is expressed in terms of the characteristic coordinate \( x = \frac{r_{\text{CM}}}{w_r} \) and \( n = \frac{N+1}{M} \). \( N \) is the vibrational quantum number of the trap associated with energy \( E_{\text{CM}} = (N+1)/(w_r^2 m_i) \). \( M \) is the angular momentum quantum number. The radial wavefunction of the valence electron with reduced mass \( \mu \) satisfies the following Schrödinger equation [52]

\[
\left[ \frac{\hbar^2}{2\mu} \frac{d^2}{d^2r} + \frac{2d}{r \, dr} + \frac{\hbar^2}{2\mu r^2} + V(r) \right] \psi_e(r)
\]

(12)

The potential \( V(r) \) is a sum of three physical contributions:

\[
V(r) = V_e(r) + V_{\text{pole}}(r) + V_{\text{so}}(r).
\]

\( V_e(r) = \frac{Z e^2}{r} \) is the Coulomb potential with \( Z \) being the effective charge from the core electron and is given by \( Z_{\text{eff}}(r) = 1 + (Z - 1) e^{-a_r r} - r (a_3 + a_4 r) e^{-a_r r} \) [52, 53]. \( V_{\text{pole}} = -\alpha_c \frac{e^2}{2r^2} \) is the potential due to the core polarization on the valence electron with \( \alpha_c \) being amplitude of the core polarisability. The values of the parameters \( a_1, a_2, a_3, a_4, r_c \) and \( \alpha_c \) can be found in standard literature [52]. Spin–orbit potential, \( V_{\text{so}} \), has the well-known form \( V_{\text{so}}(r) = \frac{\alpha}{2r^2} L \cdot S \).

Here, \( \alpha_f \) is the fine structure constant and \( (L \cdot S) = \frac{(l+1)-l(l+1)-s(s+1)}{2} \) with \( j, l \) and \( s \) being total
angular momentum, orbital angular momentum and spin angular momentum quantum number of the electron, respectively. We have solved equation (12) using the Numerov algorithm [54] to obtain the radial wavefunction. This numerical approach requires the energy values of the orbitals as input. We calculate the energy values using the quantum defect theory [55].

We consider that the atom is trapped in a two dimensional harmonic potential in the state \( n^2 S_{1/2, -1/2} \) (principal quantum number, \( n = 60 \)). For the final state \( n \) is equal to 5. For the numerical illustration of our theoretical formalism, we choose the beam waist 2.7 \( \mu \text{m} \) from the experiment of Schmiegelow et al [32]. Whereas the electric field amplitude \( (E_0) \) of the beam is considered to be 2400 \( \text{V m}^{-1} \) below the ionization limit [56] of the rubidium Rydberg atom. The characteristic length of the trap is chosen as \( w_t = 2.2 \mu \text{m} \).

We analyze our formalism with the minimum value of the OAM of the beam, i.e. \( l = 1 \). That means, according to equation (10), \( l_1 = 0 \) or 1. This study will help us to foresee the scenario for a higher OAM of the beam. Further, the first two terms in the expansion of the Gaussian factor of a LG beam (corresponding to \( q = 0 \) and 1) have been kept (see equation (A3)) to explore the radial gradient of the field. This ensures \( l_2 = 0, 1 \) and \( l_3 = 0, 1 \) in equation (11). So, with fixed values of \( l \) and \( \sigma \), we now have different transition channels depending on the quantum numbers \( l_1, l_2 \) and \( l_3 \) and they decide how the OAM from the TC and the order of the gradient of field (i.e. \( q \) or \( -q \)) are shared between the motion of the electron and the CM. Equation (10) gives the transition selection rules as,

For electron: \( \Delta L = 1 + l_1 + l_2 + l_3, 0 \leq l_1 \leq |l|, 0 \leq l_2, l_3 \leq q \) and

\[ \Delta m = l_1 + l_2 - l_3 + \sigma \]

For CM: \( \Delta M = l - l_1 - l_2 + l_3 \)

In Table 1, we show Rabi frequencies of the transitions \( S \leftarrow P \) and \( S \leftarrow D \). Finding eight possible transition channels for fixed values of TC and the polarization and calculation of Rabi frequencies considering both the CM wavefunction and the electronic wavefunction is the main novel feature of this paper. In the case of a normal atom, the Rabi frequency of the \( S \leftarrow P \) transition is normally \( 10^6 \) times stronger than that of the \( S \leftarrow D \) transition [57], but in the present case it is \( 10^2 \) times stronger. No doubt, in the interaction between a trapped Rydberg atom with the LG beam, the otherwise forbidden transitions become more prominent. This confirms the prediction of a recent paper [49]. We find that for each of these transition channels, there are two possible final states, e.g., for channel 1, the final state can be either \( P_{1/2,1/2} \) or \( P_{3/2,1/2} \), and both are equi-probable. So, instead of the \( j \) value, its projection \( m \) along the \( z \)-axis matters in transition rates only. Another important thing to note is that the different transition channels may lead to the same final state with the same transition Rabi frequencies, e.g., channel 3 and channel 5 lead to the same final state. Similarly, channel 4 and channel 6 also lead to the same final state.

Next we study the variation in Rabi frequencies for different channels. Figure 1 shows that \( S \leftarrow P \) becomes more probable as TC increases. While the \( S_{1/2, -1/2} \rightarrow D_{m=-1/2} \) transition Rabi frequency increases with TC, the \( S_{1/2, -1/2} \rightarrow D_{m=3/2} \) Rabi frequency remains more or less constant. Hence, the value of \( \Delta m \) and \( \Delta M \) are important for the transitions and they set different trends for different channels. There will be other possibilities of internal transitions here with a larger change of angular momentum of the CM, but their effects are negligible due to very weak Rabi frequencies.

| Channels | \( l_1 \) | \( l_2 \) | \( l_3 \) | \( \Delta l \) | \( \Delta m \) | Final state \( (n = 5) \) | \( \Omega \) (in KHz) |
|----------|----------|----------|----------|----------|----------|-----------------|----------------|
| 1        | 0        | 0        | 0        | 1        | 1        | \( P_{1/2,1/2} \); \( P_{3/2,1/2} \) | 607;607        |
| 2        | 0        | 0        | 1        | 2        | 0        | \( D_{3/2, -1/2} \); \( D_{5/2, -1/2} \) | 0.59;0.59      |
| 3        | 0        | 1        | 0        | 2        | 0        | \( D_{3/2,1/2} \); \( D_{5/2,1/2} \) | 1.01;1.01      |
| 4        | 0        | 1        | 1        | 3        | 1        | \( F_{3/2,1/2} \); \( F_{1/2,1/2} \) |               |
| 5        | 1        | 0        | 0        | 2        | 0        | \( D_{3/2,1/2} \); \( D_{5/2,1/2} \) | 1.01;1.01      |
| 6        | 1        | 0        | 1        | 3        | 1        | \( F_{3/2,1/2} \); \( F_{1/2,1/2} \) |               |
| 7        | 1        | 1        | 0        | 3        | 3        | \( -1 \) | \( F_{3/2,3/2} \); \( F_{1/2,3/2} \) |               |
| 8        | 1        | 1        | 1        | 4        | 2        | \( G_{7/2,3/2} \); \( G_{9/2,3/2} \) |               |

4. Conclusion

We have studied how the spatial structure of vortex light couples to both internal electronic and external CM degrees of freedom of trapped Rydberg atoms. We have treated both the electronic and the CM motions quantum mechanically. The OAM of the field can be transferred directly to the electronic motion. This leads to the modification of the standard dipole selection rule. As a result, a plethora of transition channels with different Rabi frequencies arise. Distinct identification of these channels is possible by external magnetic field. Our analysis shows that the large size of a Rydberg atom and the extended CM wavefunction lead to an appreciable effect on the Gaussian factor of the beam. This part of the interaction has been shown to be significant and on a par with the interaction arising from the inherent vorticity of the optical field. As the field OAM is shared between the electron and the CM, an entanglement induced by the LG beam is inevitable between the combined final states of the electron and the motion of the CM at the level of the \( S \leftrightarrow D \) transition.
Another outcome of this study is the generation of a mixed parity state, i.e. \( \Psi = \alpha \left| p_{/2,1/2} \right> + \beta \left| D_{/=2,3/2} \right> + \gamma \left| D_{/=2,-1/2} \right> + \ldots \). These mixing coefficients depend on the vortex charge of the LG beam. We find that the quadrupole Rabi frequency is enhanced in such an interaction. This can be experimentally verified by a procedure similar to the one discussed in [58], provided we sufficiently cool the atom below the recoil temperature limit. The change of frequency from the carrier transition to the sideband transitions, due to the linear or angular motion of the CM, will be of same order. Also, it is possible to create entanglement between internal and external angular momentum in the final states [59]. It would be interesting to carry out a similar study in the future, when the LG beam is focused [60].

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Appendix. Relations of solid harmonics

The solid harmonics \( \mathcal{R}_{\ell \ell} \) is defined by

\[
\mathcal{R}_{\ell \ell}^m = C_{\ell \ell m}^m r^m Y_{\ell \ell}^m(\theta, \phi).
\]

with \( C_{\ell \ell m}^m = \sqrt{[4\pi/(2\ell)! + 1]/(\ell - m)!/(\ell + m)!}] \).

We can express the following factors in terms of solid harmonics [61].

\[
(r \sin \theta)^{|l|} e^{i(l \phi)} = (\pm)^{|l|} \frac{[l]!}{[2|l|]} \mathcal{R}_{\ell \ell}^{|l|}(r).
\]

\[
e^{-r^2 \sin^2 \theta/\mu^2} = \sum_{q=0}^{\infty} \frac{1}{w_0^q} (2q)!^2 \mathcal{R}_{qq}^0(r) \mathcal{R}_{qq}^0(r). \quad (A3)
\]

Equations (3) and (6) show the required solid harmonics of the form \( \mathcal{R}_{\ell \ell}^m(r_{CM} + \lambda \rho) \). Additional theorems of regular solid harmonics can be used to separate internal and external coordinates as

\[
\mathcal{R}_{\ell \ell}^m(r_{CM} + \lambda \rho) = \sum_{l_i=0}^{\ell} \sum_{m_i=-l_i}^{l_i} \mathcal{C}_{l_i}^m(\lambda \rho) \mathcal{Y}_{l_i}^m(\theta_{CM}, \phi_{CM}). \quad (A4)
\]

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