Excitation of high orbital angular momentum Rydberg states with Laguerre–Gauss beams

J D Rodrigues 1, L G Marcassa 2 and J T Mendonça 1

1 Instituto de Plasmas e Fusão Nuclear, Instituto Superior Técnico, Universidade de Lisboa, 1049-001 Lisbon, Portugal
2 Instituto de Física de São Carlos, Universidade de São Paulo, Caixa Postal 369, 13560-970, São Carlos, São Paulo, Brasil

E-mail: joaodmrodrigues@tecnico.ulisboa.pt

Received 18 December 2015, revised 9 February 2016
Accepted for publication 17 February 2016
Published 17 March 2016

Abstract
We consider the excitation of Rydberg states through photons carrying an intrinsic orbital angular momentum degree of freedom. Laguerre–Gauss modes, with a helical wave-front structure, correspond to such a set of laser beams, which carry \( \ell_0 \) units of orbital angular momentum in their propagation direction, with \( \ell_0 \) the winding number. We demonstrate that, in a proper geometry setting, this orbital angular momentum can be transferred to the internal degrees of freedom of the atoms, thus violating the standard dipole selection rules. Higher orbital angular momentum states become accessible through a single photon excitation process. We investigate how the spacial structure of the Laguerre–Gauss beam affects the radial coupling strength, assuming the simplest case of hydrogen-like wavefunctions. Finally we discuss a generalization of the angular momentum coupling, in order to include the effects of the fine and hyperfine splitting, in the context of the Wigner–Eckart theorem.

Keywords: Rydberg atoms, Laguerre–Gauss beams, selection rules

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

1. Introduction

Rydberg atoms [1–5] are amongst the most exciting and promising research topics nowadays. Much of this interest arises from the particularities of these systems, such as strong dipole–dipole interactions and long lifetimes. The dipole blockade [6–9] is an example of the implications of strong interactions and, applications to quantum information [3] were proposed based on possibility of performing quantum gate operations based on this effect [10–12]. These unique features become even more evident when we consider the propagation of light in Rydberg gases. Particularly, photon–photon interactions can be tailored in the blockade regime, under the conditions of electromagnetically induced transparency [13], leading, for instance, to the formation of Coulomb-like bound states of photons [14].

Until now and in most experiments, the Rydberg atoms were excited using dipole allowed transitions, either from the ground state or from an intermediate excited state. Only recently, electric dipole-forbidden transitions to Rydberg \( nD \) states were observed in Rubidium atoms [15]. Molecular resonances involving two Rydberg atoms, also called microdimers, were observed and associated with quadrupole–quadrupole interactions [16, 17]. A dipole–quadrupole molecular resonance was observed recently by Deiglmayr and co-workers in Caesium [18]. All these experiments have something in common, they require a high intensity laser beam to observe such transitions since their rates are very small. It would be interesting to increase such rate in order to manipulate the properties of Rydberg atom interactions. In a recent work, Schmiegelow and co-workers have used Laguerre–Gauss (LG) laser beams to study a quadrupole transition in a Calcium ion [19]. In principle such a technique could be applied to cold Rydberg atoms as well.

In this work we investigate the excitation of Rydberg states using orbital angular momentum (OAM) carrying photons, namely with LG laser beams. As Allen and co-workers discovered in 1992, LG light fields carry a discrete amount of orbital angular momentum per photon [20, 21]. LG photons have been employed to encode quantum information...
atom, which captures the essential ingredients of the OAM. In section 4, we discuss the nature of the angular momentum coupling between the LG photon and the atom in a hyperfine structure basis. Finally, in sections 5 and 6, we discuss the results and possible applications and state some conclusions.

2. Dipole selection rules

Photon excitation processes usually rely on the interaction of the atomic dipole moment, \( \mu = -e \mathbf{r} \), where \( \mathbf{r} \) is the distance of the electron to the positively charged nucleus, and the electric field of a laser beam, \( \mathbf{E} = e E_0 S(\mathbf{r}) e^{i\omega t} \). In this expression \( e \) is the unit polarization vector of the laser field and \( S(\mathbf{r}) \) the spatial and phase structure of the beam, as determined by the Helmholtz equation
\[
(\nabla^2 + k^2) S(\mathbf{r}) = 0,
\]
with wavenumber \( k \).

2.1. Plane-wave excitation

Plane waves correspond to the particular solution of equation (1) \( S(\mathbf{r}) = e^{-i k \mathbf{r}} \). In the usual dipole approximation, \( e^{-i k} \approx 1 \), we simply have \( e \approx e_0 e^{i \omega t} \), where the size of the atomic ground state is much smaller than the photon wavelength, \( r_0 \ll \lambda \), so that the atom only senses a temporal evolution of the field. In first order perturbation theory, this time-dependent interaction, with Hamiltonian \( H_{\text{int}} = \mu \cdot \mathbf{E} \), couples the eigenstates of the free Hamiltonian, and the relevant quantity is the dipole matrix element defined as
\[
\mu = -e \langle \psi | e \cdot \mathbf{r} | \psi \rangle = -e \langle nm\ell | e \cdot \mathbf{r} | n'\ell' m' \rangle,
\]
where the initial and final quantum states are specified by the usual quantum numbers \( n, \ell \) and \( m \). This is reasonable for hydrogen-like atoms. Later, a more accurate description of the atomic state will be given. It becomes advantageous to represent the laser polarization on a spherical basis as \( e = e_{-1} e_{-1} + e_{0} e_{0} + e_{1} e_{1} \), with \( e_{\ell} = 1/\sqrt{2} (e_{\ell} \pm i e_{-\ell}) \) and \( e_{0} = e_{-0} \). Note that \( (e_{\ell}, e_{0}) \) are associated with left-circularly, linear and right-circularly polarized light. In this basis, the dipole operator can be written as
\[
\mu_q = -e r \sqrt{4\pi/3} Y_1^q (\theta, \phi) \text{ with } Y_1^q (\theta, \phi) \text{ the spherical harmonic with } \ell = 1 \text{ and } m = q, \text{ corresponding to the various photon polarization states such that } q = (-1, 0, +1) \text{ is associated with } (\sigma_-, \sigma_0, \sigma_+) \text{ transitions. The transition matrix element in equation (2) can then be written as}
\]
\[
\mu_q = \langle nm\ell | -e r \sqrt{4\pi/3} Y_1^q | n'\ell' m' \rangle = \frac{4\pi}{3} \langle m|Y_1^q|n'\ell' m'| \rangle - e r \langle n'\ell' m' \rangle.
\]

The angular coupling in the first term of this expression is given in terms of Clebsch–Gordan coefficients
\[
\langle n|Y_1^q|n'\ell' m' \rangle = \frac{(2\ell + 1)(2L + 1)}{4\pi (2\ell' + 1)} \mathcal{C}_{L,M,L'}^{\ell,\ell'} \mathcal{C}_{0,0,0}^{LL',MM'},
\]
where \( L, M, L' \) and \( M' \) are the total angular momentum and the projections of the total angular momentum respectively.
determining the strength of the angular momentum coupling, in this case \((\ell, m) \rightarrow (\ell', m')\). From the properties of the Clebsch–Gordan coefficients for angular momentum addition we can easily realize that for such a process as the one in equations (2) and (3), the angular coupling results in 
\[ \Delta m = \pm 1, 0 \quad \text{and} \quad \Delta \ell = \pm 1, \]
the usual dipole selection rules, with \(\Delta m = m' - m\) and \(\Delta \ell = \ell' - \ell\).

2.2. LG excitation

Let us now consider the case where the atomic excitation is achieved using a LG beam in the geometry setting described in figure 1. Such modes appear as solutions of the Helmholtz equation [1] in the paraxial approximation. To capture the essence of this approximation let us write the structure function as \(S(r) = s(r_z) e^{-ikz}\). Under the condition of strong paraxial propagation, namely \(|\partial^2 s/\partial z^2| < |k\partial s/\partial z|\), corresponding to a slowly-varying amplitude function \(s(r_z)\) along propagation direction \(z\), the Helmholtz equation reduces to

\[
\left(\nabla^2 + 2ik \frac{\partial}{\partial z}\right)s(r_z) = 0. \tag{5}
\]

In this case, the structure function \(S(r)\), in the dipole approximation and for a given \((p_0, \ell_0)\) mode, is determined by [37, 38]

\[
S_{p_0\ell_0}(r) = C_{p_0\ell_0} \varsigma(p_0) L_{p_0}^{\ell_0}(\zeta) e^{-\zeta^2/2} e^{i\ell_0\phi}, \tag{6}
\]

with the integers \(p_0\) and \(\ell_0\) being the radial and azimuthal mode numbers, respectively, \(\zeta \equiv \sqrt{2} \rho/w_0\) and \(w_0\) the beam waist. The normalization constants \(C_{p_0\ell_0}\) are given by

\[
C_{p_0\ell_0} = \left(\frac{2p_0!}{\pi^{1/4} \ell_0! p_0^{1/2}}\right)^{1/4}, \quad \text{and the usual Laguerre polynomials}
\]

\[
L_{p_0}^{\ell_0}(x) = \frac{e^x}{p_0^{1/2} \sqrt{\pi}} \frac{d^p_0}{dx^p_0} \left[x^{p_0} e^{-x}\right]. \tag{7}
\]

Note that we are ignoring the Gouy phase factor, and the variation of the beam waist and the beam curvature radius along the \(z\) direction, which is reasonable in the dipole approximation. Moreover, these corrections are not important to the effects we are trying to capture here. The winding number, \(\ell_0\), defines the amount of OAM carried by each photon, in this case \(\ell_0\). Typically, the spread of ground state wavefunction is much smaller than the beam waist and we can approximate the dipole matrix element as

\[
\mu = -e \langle \psi_f | e \cdot r \left(\frac{2\sqrt{r} \sin \theta}{w_0}\right) | \ell_0 \rangle e^{i\ell_0\phi} \langle \psi_f \rangle \tag{8}
\]

with \(r \sin \theta = \rho\). In the same way as before, \(e \cdot r = r \sqrt{4\pi/3} Y_i^\ell\). The angular momentum properties of the LG beam are encoded in the term \(\sin \theta e^{i\ell_0\phi}\) which can be written in terms of spherical harmonics as \(\sin \theta e^{i\ell_0\phi} = \left(2\sqrt{\frac{2\pi}{3}}\right)^{1/4} Y_{\ell_0}^{\ell_0\ell_0}(\theta, \phi) \) with \(\text{sgn}(\phi)\) the sign function. Let us now rewrite the matrix element in equation (8) as

\[
\mu = -e \left(\frac{8}{3!\ell_0!}\right)^{1/4} \langle \ell m | Y_i^\ell | \ell' m' \rangle \times \langle n\ell | \left(\frac{2\sqrt{r}}{w_0}\right) | n'\ell' \rangle, \tag{9}
\]

where we separated the matrix element into angular and radial couplings. We shall now explore the nature of the object \(Y_i^\ell | Y_{\ell_0}^{\ell_0\ell_0}(\theta, \phi) \rangle \). Let us begin with the OAM term \(Y_i^\ell | Y_{\ell_0}^{\ell_0\ell_0}(\theta, \phi) \rangle \), which corresponds to the \(|\ell_0\rangle\) power of the simple spherical harmonic \(Y_i^\ell\). Using the known result for the multiplication of spherical harmonics (see appendix A) we obtain

\[
\left(2\sqrt{\frac{2\pi}{3}}\right)^{1/4} \langle \ell m | Y_i^\ell | \ell_0 \rangle | 0 \rangle = 2\ell_0! |\ell_0\rangle \left(\frac{4\pi}{(2\ell_0 + 1)!}\right) Y_i^{\ell_0}(\theta) \tag{10}
\]

which, apart from a normalization factor, simply corresponds to an angular momentum state with \(\ell = |\ell_0|\) and \(m = \ell_0\), as expected. Let us now focus on the joint effect of the photon polarization and orbital angular momentum by investigating the full operator \(Y_i^\ell | Y_{\ell_0}^{\ell_0\ell_0}(\theta, \phi) \rangle \). Ignoring for now the normalization constants, for the sake of simplicity, and proceeding in the same way as before (see appendix B) we can write

\[
Y_i^{\ell_0}(\theta) = \sum_{\ell'} \sum_{m} m(q, \ell_0, \ell') Y_i^{\ell_0+\ell'}(\theta), \tag{11}
\]

with

\[
m(q, \ell_0, \ell') = \frac{3(2|\ell_0| + 1)}{4\pi (2\ell_0 + 1)!} C_{q,0,q+\ell}^{\ell_0,0,0} C_{\ell_0,0,0}^{\ell_0,0,0} \tag{12}
\]

where the sum is performed over \(\max\{|\ell_0| - 1, |\ell_0 + q|\} \leq \ell' \leq |\ell_0| + 1\), so that the Clebsch–Gordan coefficient in equation (12) is non-vanishing. Putting together equations (9)–(12) allows us to write

\[
\mu = -e D(\ell_0) \mathcal{R}(n, n', \ell, \ell', 0) \times \sum_{\ell'} \sum_{m} m(q, \ell_0, \ell') \langle \ell m | Y_i^{\ell+\ell'} | \ell' m' \rangle \tag{13}
\]

with \(D(\ell_0) = 2|\ell_0| \sqrt{|\ell_0| + 1/2 |\ell_0| + 1/2 + 1} \),

\[
\mathcal{R}(n, n', \ell, \ell', 0) = |n\ell | \left(\frac{\sqrt{2\pi}}{w_0}\right) \left| n'\ell' \right| \tag{14}
\]

the radial coupling, modified by the geometry of the LG beam. Combining equations (4) and (13) we can finally write

\[
\mu = -e D(\ell_0) \mathcal{R}(n, n', \ell, \ell', 0) \times \sum_{\ell'} \mathcal{M}_i^{\ell} (q, \ell_0, \ell') C_{\ell_0,0,0}^{\ell_0,0,0} \tag{15}
\]

with

\[
\mathcal{M}_i^{\ell} (q, \ell_0, \ell') = m(q, \ell_0, \ell') \sqrt{(2\ell_0 + 1)(2\ell_0 + 1) + 1} \ C_{\ell_0,0,0}^{\ell_0,0,0}. \tag{16}
\]

It is the angular coupling associated with the Clebsch–Gordan coefficients in equation (15) that determines the selection rules of the excitation process described here. It directly
follows from the conservation law for the sum of angular momentum projections that, a non-zero coefficient implies

$$\Delta m = m' - m = \ell_0 + q.$$  

(17)

Another important condition arising from the symmetry properties of the Clebsch–Gordan coefficients is

$$|\ell - \ell'| \leq \ell' \leq \ell + \ell''.$$  

(18)

This relation is known as the triangular condition. Note that a third imposition arises due to the parity properties of the spherical harmonics. The wave-function of the state $|\ell m\rangle$ has parity $P(Y^\ell_m) = (-1)^\ell$ and, the condition of an overall (-1) parity in each of the matrix element integrals implies that

$$\Delta \ell + |\ell''|$$  

is even.  

(19)

In the above expressions $\ell''$ ranges between $\max(|\ell_0| - 1, |\ell_0 + q|) \leq \ell'' \leq |\ell_0| + 1$. Together, equations (17)–(19) define the dipole selection rules for the excitation process described here. If it often important, given an initial atomic state with OAM quantum number $\ell$, to understand which are the open transitions in the final $\ell'$ quantum number, independent of the atomic initial and final magnetic quantum numbers, $m$ and $m'$ respectively. In this sense we can define the quantity

$$A_{q,m}(\ell, \ell_0, \ell') = \sum_{q,m} |D(\ell_0)| \sum_{\ell''} M^{(q)}_{\ell''}(q, \ell_0, \ell'')$$

$$\times e^{i\ell \ell' \ell'' + \ell_0 m + q + \ell_0 q + \ell'},$$  

(20)

where we used the selection rule in equation (17) to eliminate the dependence on initial and final magnetic quantum numbers, as well as in the photon polarization state $q$. In figure 2 we plot such a function, for the case of an initial $P$ orbital. In the case of $^{85}$Rb, for instance, the Rydberg excitation process usually involves a two photon process. In the first step we excite the ground state, $5S_{1/2}$, to the excited $5P_{3/2}$ state, corresponding to the D2 line at approximately 780 nm. A second pulse can then be used to the excitation of high-lying Rydberg states.

3. Modified radial coupling

In this section we will numerically investigate the radial coupling, $R$, as defined in equation (14) and, in particular, how it is modified by the transverse structure of the LG modes. For this purpose we will make use of the hydrogen atom wavefunctions, which are a good approximation for the alkali metals. In this case, analytical solutions for the normalized radial wavefunctions exist, and are given by [39]

$$R_{\ell d}(r) = -\frac{2}{(n \ell + 1)!} \frac{2r}{2n} \frac{2\ell + 1}{2n!} \frac{2\ell + 1}{2n!} e^{-r^2/2n^2} L^{2\ell+1}_{2n+1}(r^2),$$  

(21)

with $a_0$ the Bohr radius and $r^* = 2r/a_0$. Later we shall discuss the validity of this approximation. In figure 3 we compute the radial coupling for different sets of photon and final atomic angular momentum states. The dependence on the principal quantum number $n$ is plotted and the scaling is investigated by fitting the numerical results to a function of the type $f(n) = C_\ell n^p$. We obtain $p = -3.2, -3.3, -2.8$ and $-2.4$, for $\ell_0 = 0, \ell_0 = 1, \ell_0 = 2$ and $\ell_0 = 3$, respectively. Note that, in the usual case of a plane wave excitation, $\ell_0 = 0$, the phenomenological scaling evidenced in experimental measures corresponds to $p = -3$ [40]. Clearly, for higher photon OAM, the rescaled matrix element in figure 3 also increases significantly, due to the higher powers of the electron position operator, $x^{2\ell+1}$. Nevertheless, to obtain the important quantity relevant for experiments, we shall multiply the rescaled matrix element by the ratio of the Bohr radius to the beam waist, $\sqrt{2a_0}/w_0$.

The hydrogen wavefunctions used in the present discussion correspond to a first approximation when dealing with alkali metals, where the valence electron is immersed in
a \mathcal{a} - e/r \text{ Coulomb potential as the closed electron shells screen the nuclear charge. Nevertheless, for lower angular momentum states, typically } \ell \leq 3, \text{ the electron wavefunction penetrates these closed shells and becomes exposed to the unscreened nuclear charge. This causes a deviation from the pure Coulombic potential at shorter ranges, which can be accounted for with the introduction of the quantum defect } \delta_{eq}\text{, and an effective principal quantum number as } n^* = n - \delta_{eq} [1]. \text{ For high orbital angular momentum states, typically } \ell > 3, \text{ the potential is, to a high level of accuracy, a pure Coulomb potential and the quantum defects are zero. The simplified model used here allow us to capture the essentials for the effects we are trying to capture.}

4. Generalized angular coupling

We have so far assumed the atomic state to be completely described by the angular momentum quantum numbers \( \ell \) and \( m \) (along with the principal quantum number \( n \)). This allowed to tackle the problem in a simplified manner, without considering the effects of spin–orbit coupling, responsible for the fine-structure splitting of the energy levels as \( J = L + S \). This total electron angular momentum \( J \) also couples with the nucleus angular momentum \( I \) as \( F = J + I \), resulting in the hyperfine structure of the atomic levels. In this case, an atomic state is completely described by a larger set of quantum numbers as \( |m(s)j(i)m_f, I, J, L, S, \ell, m, \ell, m_{f}' \rangle \), where the spin and nuclear angular momentum quantum numbers, \( s \) and \( i \), respectively, are constant and do not couple with the photon. The question now arises of how to integrate these effects on the analysis develop so far. The answer is given by the Wigner-Eckart theorem, as it explores the symmetry properties of the Wigner-Eckart theorem, which re-earlier, and dictated by the symmetry properties of the Clebsch-Gordan coefficients, can alternatively be determined by the Wigner-3j symbols

\[
\langle \ell(s)j(i)f m_f|\mathcal{Y}_\ell^m|\ell'(s)j'(i)f' m_{f}' \rangle = \sum_{\ell''} m(q, \ell_0, \ell'') \langle \ell(s)j(i)f m_f|\mathcal{Y}_{\ell''}^{m''}|\ell'(s)j'(i)f' m_{f}' \rangle.
\]

where the angular coupling now depends on a larger set of quantum numbers. Note that the selection rules derived earlier, and dictated by the symmetry properties of the Clebsch–Gordan coefficients, can alternatively be determined by the Wigner-3j symbols

\[
\begin{pmatrix} \ell & L & \ell' \\ m & M & -m' \end{pmatrix} = (-1)^{l'-l-m'} \frac{1}{\sqrt{2l'+1}} C_{m,M,m'}^{\ell,L,\ell'},
\]

which reflect the same angular momentum conservation properties. The objects \( \mathcal{Y}_\ell^m \) constitute a set of spherical tensor operators, satisfying the commutation relations

\[
[J_\ell, \mathcal{T}_k^m] = q \mathcal{T}_k^m \quad \text{and}
\]

\[
[J_\ell, \mathcal{T}_k^m] = \sqrt{k(k+1)} - q(q \pm 1) \mathcal{T}_k^{m\pm 1}.
\]

In this way, we can make use of the Wigner–Eckart theorem to write [41, 42]

\[
\langle \ell(s)j(i)f m_f|\mathcal{Y}_\ell^m|\ell'(s)j'(i)f' m_{f}' \rangle = (-1)^{m'} \times \left\{ \begin{array}{c} f \ \ell^+ \ f' \\
 m_f \ q + \ell_0 - m_f' \end{array} \right\} \langle \ell(s)j(i)|\mathcal{Y}_\ell'|\ell'(s)j'(i)f' \rangle,
\]

where we have factorized the dependence on the magnetic quantum number \( m_f \) inside the Wigner-3j symbol and introduced the reduced matrix element, independent of the photon polarization state \( q + \ell_0 \). In the same way as before, given the properties of the Wigner-3j symbols, a non-vanishing coefficient implies

\[
\Delta m_f = m_f' - m_f = \ell_0 + q,
\]

which is the natural generalization of the selection rule in equation (17). We shall now notice that, since the photon does not couple with the nuclear angular momentum, \( I \), we can factorize its dependence on the reduced matrix element in equation (26) into a Wigner-6j symbol as [43]

\[
\langle \ell(s)j(i)f|\mathcal{Y}_\ell|\ell'(s)j'(i)f' \rangle = (-1)^{j+j'+1+i} \times \sqrt{2(f' + 1)(2j + 1)} \left\{ \begin{array}{c} j \ j' \ 1 \\
 f' \ f \ i \end{array} \right\} \langle \ell(s)j|\mathcal{Y}_\ell|\ell'(s)j' \rangle.
\]

In the same way, given that the spin of the electron, \( S \) does not change, we can further factorize the reduced matrix element as

\[
\langle \ell(s)j|\mathcal{Y}_\ell|\ell'(s)j' \rangle = (-1)^{j'+\ell+1+i} \times \sqrt{2(j'+1)(2\ell+1)} \left\{ \begin{array}{c} \ell \ \ell' \ 1 \\
 j \ j' \ s \end{array} \right\} \langle \ell|\mathcal{Y}_\ell|\ell' \rangle,
\]

where we have defined the reduced matrix element

\[
\langle \ell|\mathcal{Y}_\ell|\ell' \rangle = \frac{\sqrt{2(\ell+1)(2\ell^*+1)(2\ell'+1)}}{4\pi} \left( \begin{array}{c} \ell \ \ell^* \ \ell' \end{array} \right).
\]

Note that, by setting \( I = 0 \) and \( S = 0 \) in equation (28) and (29) we recover the previous results. Moreover, note that the result in equation (30) allows us to recover the selection rules for quantum number \( \ell \) derived in section 2, as it corresponds to the useful information when determining the accessible quantum states. In fact, and as stated before, since the electron spin and the nucleus angular momentum do not couple to the photon, the results presented in this section only introduce a correction on the angular coupling strength of the allowed transitions.

5. Discussion

We shall now discuss some of the implications and limitations of the analysis develop so far. We have shown that the OAM carried by an LG photon can be transferred to the internal degrees of freedom of the atom, as suggested by recent experimental results [19]. We considered the case where the
atomic ground state is centred within the optical vortex. In fact, as long as the optical vortex is contained within the spacial extent of the ground state wavefunction, such a transfer of OAM should be expected, broadening the range of accessible quantum states as dictated by the new selection rules in equations (17)–(19).

Note that, in the case of a plane wave excitation, dipole-forbidden transitions can occur at much lower rates. For instance, the dipole-forbidden 5S → nD, with n = 27 ~ 59, is observed via a one-photon quadrupole transition, although at a rate that is lower by a factor of approximately 2000, when compared to the dipole-allowed transition 5S → nP, in Rubidium atoms [15]. On the other hand, by using an LG beam with winding number ℓ₀ = 1 and beam waist ω₀ ~ 1 μ m, in the geometrical setting described earlier, both the atomic transitions 5S → nD and 5P → nP became allowed and, moreover, we can expect them to occur at a rate which is only 10 to 100 times lower than the transition rate for 5S → nP and 5P → nD with a usual plane wave excitation, assuming the same electric field peak intensity E₀.

The main drawback here is the vanishing of the electric field amplitude at the optical vortex and, as such, focusing the excitation beam close to the diffraction limit shall be important. Moreover, the displacement of the atom from the optical vortex will reduce the probability of OAM transfer [44]. The excitation could be performed in an atomic sample trapped in an optical dipole trap, using a combination of two perpendicular focused laser beams. In the case of Rubidium, for instance, a 780 nm laser could excite the Rydberg state. This new excitation could be performed in a atomic sample trapped in an optical dipole trap, using the only non-vanishing term in the sum is ℓ = 2 and we simply have

\[ Y_1^2 Y_1^2 = \frac{(1, 2, 0, 0)}{4\pi (5)} Y_1^0 Y_1^0 \]  

with \( C_{1,1,2}^{1,1,2} = 1 \) and \( C_{0,0,0}^{1,1,2} = \sqrt{2/3} \). Notice that, when computing a n-th power of the spherical harmonic \( Y_1^i \), the terms \( C_{n-1-i,1,n-i}^{n-1-i,1,n-i} \) and \( C_{0,0,0}^{1,1,2} \) appear in the product, with \( i = 0, \ldots, n - 2 \). In this sense, we shall notice that

\[ C_{n-1-i,1,n-i}^{n-1-i,1,n-i} = 1, \quad C_{0,0,0}^{1,1,2} = \frac{\ell + 1}{2\ell + 1}. \]  

All the above analysis remains valid for powers of the spherical harmonics \( Y_1^i \). From equation (A1) and the properties in equation (A3) we can write

\[ [Y_1^{\text{sgn}(\ell_1)}]_{\ell_1} = \prod_{i=1}^{[\ell_1]} \frac{3(i + 1)}{4\pi (2i + 3)} Y_1^{\ell_1} \]  

We can now transform this particular multiplication into an expression involving factorial terms, namely

\[ 2 \sqrt{\frac{2\pi}{3}} Y_1^{\text{sgn}(\ell_1)} \prod_{i=1}^{[\ell_1]} \frac{4\pi}{2(\ell_1 + 1)!} Y_1^{\ell_1} \]  

**Appendix B**

Let us compute the joint effect of photon polarization and orbital angular momentum, determined by the operator

\[ J D R \text{ acknowledges the financial support of FCT—Fundação para a Ciência e Tecnologia (Portugal) through the grant number SFRH/BD/52323/2013. This work was partially supported by grant 2013/02816-8, São Paulo Research Foundation (FAPESP), AFOSR (FA9550-12-1-0434), U.S. Army (W911NF-15-1-0638) and CNPq.} \]
as before, we start with the expression for the product of spherical harmonics in equation (A1). In this case we are left with

\[ Y_l^q Y_l^{q_0} = \sum_{\ell} \frac{3(2|\ell_0| + 1)}{4 \pi (2\ell + 1)} C^{1}_{|\ell_0|,\ell_0,\ell_0} C^{1}_{0,0,0,0} Y_{\ell}^{q+q_0}, \]  

(B1)

with \( \max \{||\ell_0| - 1|, |\ell_0 + q_0| \leq \ell \leq |\ell_0| + 1 \). For the sake of simplicity we write

\[ Y_l^q Y_l^{q_0} = \sum_{\ell} \frac{3(2|\ell_0| + 1)}{4 \pi (2\ell + 1)} C^{1}_{|\ell_0|,\ell_0,\ell_0} C^{1}_{0,0,0,0} Y_{\ell}^{q+q_0} \]  

(B2)

with

\[ m(q, \ell_0, \ell) = \frac{3(2|\ell_0| + 1)}{4 \pi (2\ell + 1)} C^{1}_{|\ell_0|,\ell_0,\ell_0} C^{1}_{0,0,0,0}. \]  

(B3)

References

[1] Gallagher T F 1994 Rydberg Atoms (Cambridge: Cambridge University Press)
[2] Marcassa L G and Shaffer J P 2014 Adv. At. Mol. Opt. Phys. 63 47
[3] Saffman M, Walker T G and Molmer K 2010 Rev. Mod. Phys. 82 2313
[4] Low R, Weimer H, Nipper J, Balewski J B, Butscher B, Buchler H P and Pfau T 2012 J. Phys. B: At. Mol. Opt. Phys. 45 113001
[5] Cabral J S et al 2011 J. Phys. B: At. Mol. Opt. Phys. 44 184007
[6] Comparat D and Pfau T 2010 J. Opt. Soc. Am. B 27 A208
[7] Weidemuller M 2009 Nat. Phys. 5 91
[8] Urban E, Johnson T A, Henage T, Isenhower L, Yavuz D D, Walker T G and Saffman M 2009 Nat. Phys. 5 110
[9] Gallagher T F and Pfau T 2008 Adv. At. Mol. Opt. Phys. 56 161–218
[10] Jaksch D, Cirac J I, Zoller P, Rolston S L, Côté R and Lukin M D 2000 Phys. Rev. Lett. 85 2208
[11] Lukin M D, Fleischhauer M, Cote R, Duan L M, Jaksch D, Cirac J I and Zoller P 2001 Phys. Rev. Lett. 87 037901
[12] Maller K M, Lichtman M T, Xia T, Sun Y, Piotrowicz M J, Carr A W, Isenhower L and Saffman M 2015 Phys. Rev. A 92 022336
[13] Gorshkov A V, Otterbach J, Fleischhauer M, Pohl T and Lukin M D 2011 Phys. Rev. Lett. 107 133602
[14] Maghrebi M F, Gullans M J, Bienias P, Choi S, Martin I, Firstenberg O, Lukin M D, B"uchler H P and Gorshkov A V 2015 Phys. Rev. Lett. 115 123601
[15] Tong D, Farooqi S M, van Kempen E G M, Pavlovic Z, Stanojevic J, Côté R, Eyler E E and Gould P L 2009 Phys. Rev. A 79 052509
[16] Stanojevic J, Côté R, Tong D, Eyler E E and Gould P L 2008 Phys. Rev. A 78 052709
[17] Schwettmann A, Crawford J, Overstreet K R and Shaffer J P 2006 Phys. Rev. A 74 020701
[18] Deiglmayr J, Salomaa-Mausknas H, Pillet P and Merkt F 2014 Phys. Rev. Lett. 113 193001
[19] Schmiegelow C T, Schulz J, Kaufmann H, Ruster T, Poschinger U G and Schmidt-Kaler F 2015 (arXiv:1511.07206)
[20] Allen L, Beijersbergen M W, Spreeuw R J C and Woerdman J P 1992 Phys. Rev. A 45 8185
[21] Beijersbergen M, Allen L, van der Veen H and Woerdman J P 1993 Opt. Commun. 96 123
[22] Fickler R, Lapkiewicz R, Huber M, Lavery M P J, Padgett M J and Zeilinger A 2014 Nat. Comm. 5 article 184007
[23] Nogues L, Sansoni L, Scarianno F, De Martini F, Marrucci L, Piccirillo B, Karimi E and Santamato E 2009 Nat. Photon. 3 720
[24] Souza C E R, Borges C V S, Khoury A Z, Huguenin J A O, Aolita L and Walborn S P 2008 Phys. Rev. A 77 032345
[25] Nicolas A, Veissier L, Giner L, Giacobino E, Maxein D and Laurat J 2014 Nat. Photon. 8 234 letter
[26] Beijersbergen M, Coerwinkel R, Kristensen M and Woerdman J 1993 Opt. Commun. 112 321
[27] Curtis J E, Ross B A and Grier D G 2002 Opt. Commun. 207 169
[28] Mirhosseini M, na Loaiza O S M, Chen C, Rodenburg B, Malik M and Boyd R W 2013 Opt. Express 21 30196
[29] Franke-Arnold S, Allen L and Padgett M 2008 Laser Photon. Rev. 2 299
[30] Yao A M and Padgett M J 2011 Adv. Opt. Photon. 3 161
[31] Andersen M F, Ryu C, Cladé P, Natarajan V, Vaziri A, Helmerson K and Phillips W D 2006 Phys. Rev. Lett. 97 170406
[32] Inoue R, Kanai N, Yonohara T, Miyamoto Y, Koashi M and Koszuma M 2006 Phys. Rev. A 74 053809
[33] Mendonca J T, Thidé B and Then H 2009 Phys. Rev. Lett. 102 180505
[34] Vieira J and Mendonca J T 2014 Phys. Rev. Lett. 112 215001
[35] Picon A, A B, Mompart J, Vazquez de Aldana J R, Plaja L, Calvo G F and Roso L 2010a New J. Phys. 12 083053
[36] Picon A, Mompart J, de Aldana J R V, Plaja L, Calvo G F and Roso L 2010b Opt. Express 18 3660
[37] Bond C, Fulda P, Carbone L, Koeckermann K and Freise A 2011 Phys. Rev. D 84 102002
[38] Klimov V V, Bloch D, Duloy M and Rios Leite J R 2012 Phys. Rev. A 85 053834
[39] Schiff L 1949 Quantum Mechanics (New York: McGraw-Hill Book Company)
[40] Deiglmayr J, Reetz-Lamour M, Amthor T, Westermann S, de Oliveira A and Weidemuller M 2006 Opt. Commun. 264 293
[41] Wigner E 1959 Group Theory and Its Application to the Quantum Mechanics of Atomic Spectra (New York: Academic Press)
[42] Brink D and Satchler G 1962 Angular Momentum (Oxford)
[43] Edmonds A 1968 Angular Momentum in Quantum Mechanics (Princeton University Press)
[44] Afanasev A, Carlson C E and Mukherjee A 2013 Phys. Rev. A 88 033841
[45] Nascimento V A, Caliri L L, Schwettmann A, Shaffer J P and Marcassa L G 2008 Phys. Rev. Lett. 102 213201
[46] Cabral J S, Kondo J M, Onaças J L, Marcassa L G, Booth D, Tallant J and Shaffer J P 2010 New J. Phys. 12 093023
[47] Nascimento V A, Caliri L L, de Oliveira A L, Bagatto V S and Marcassa L G 2006 Phys. Rev. A 74 054501