Quadrupole absorption rate and orbital angular momentum transfer for atoms in optical vortices

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Recent experiments involving the interaction of optical vortices with atoms in quadrupole transitions have shown it to be accompanied by the exchange of orbital angular momentum (OAM) between the electronic states of the atom and the optical vortex field. Earlier work, both theoretical and experimental, had ruled out the transfer of a vortex OAM to the electronic degrees of freedom in an electric dipole atomic transition and it has been confirmed that the lowest multipolar order involving an OAM transfer to the electronic motion is indeed the electric quadrupole. Hitherto, the quadrupole transition involving optical vortices has not been quantified and we thus set out to evaluate the absorption rate accompanied by an OAM transfer with reference to the $6\,^2S_{1/2} \rightarrow 5\,^2D_{5/2}$ in Cs when cesium atoms are subject to the field of a linearly polarized optical vortex. Our results assuming typical experimentally accessible parameters indicate that the absorption rate for moderate light intensities is lower than the quadrupole spontaneous emission rate but should still be within the measurement capabilities of modern spectroscopic techniques.

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I. INTRODUCTION

Twisted light beams or optical vortices have been the subject of much investigation over the last three decades or so and this area has found applications in a number of diverse fields [1–3]. The absorption or emission of any type of light by atoms is accompanied by the transfer of linear momentum between the light and the atoms, and this effect has been exploited in the cooling and trapping of atoms [4–6]. It has also been established that the application of vortex light to atoms affects the center-of-mass motion [6,7] via the dipole force, but the quadrupole force too can influence atomic motion attracting the atoms to high- or low-intensity regions of the light field [8–10].

The possibility of the exchange of orbital angular momentum (OAM) between light and the internal motion of atoms, as opposed to their gross, or center-of-mass, motion, has interested researchers since the early 1990s [11,12]. The first treatments to tackle the problem of transfer [13] concluded that while the center of mass of atoms engages with an optical vortex, the internal 'electronic-type' degrees of freedom of the atom do not take part in any OAM exchange in an electric dipole transition and that only in a quadrupole interaction can an exchange involving the electronic (internal) degrees of freedom take place. This result was subsequently confirmed experimentally by a number of researchers, first by Araoka *et al.* [14], who showed that optical vortex light is not specific in the interaction with chiral matter, and then by Löffler *et al.* [15,16], whose experimental work did not detect any influence of the OAM of circular dichroism in cholesteric polymers. The recent experimental work by Giammanco *et al.* [17] confirmed categorically the lack of influence of the OAM on electric dipole transitions, in agreement with theory [13,14]. The experimental work by Schmiegelow *et al.* [18] has shown that an atom or an ion can exchange two units of optical angular momentum, one unit from optical spin and another from its OAM. Most previous studies have dealt primarily with the case in which the optical vortex light is linearly polarized and so optical spin has no role to play in the transfer process.

Research has also dealt with the study of the mechanical forces on atoms due to the coupling of optical vortices, such as the Laguerre-Gaussian (LG) and Bessel-Gaussian modes, to quadrupole-active atomic transitions [6,10,19] and the results displayed a considerable enhancement in the case of twisted beams due to the gradient coupling, which increases with increasing winding number. Quadrupole transitions have also been observed in the case of Rb atoms by evanescent light when Rb is localized in the vicinity of an optical nanofiber [9].

As potentially measurable effects the absorption of linearly polarized vortex light and the rate of absorption by atoms have not been evaluated as far as the authors are aware. We have therefore set out to evaluate the rate of absorption, which in the case of an optical vortex is also interpretable as the rate of OAM transfer from the vortex light to the atoms. The general treatment is then applied to the particular case involving the $6\,^2S_{1/2} \rightarrow 5\,^2D_{5/2}$ quadrupole transition in Cs when cesium atoms are subject to the field of an optical vortex. The Cs transition in question is well known as a dipole-forbidden but quadrupole-allowed transition.

This paper is organized as follow. The basic theory is outlined in Sec. II, while Sec. III presents the theory of the quadrupole atomic absorption rate when the atom engages

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with the optical vortex field at near-resonance. The evaluation involves a direct application of the Fermi Golden Rule, where the usual selection rules are applicable to quadrupole transitions, but absorption requires a treatment involving the density of the continuum states as a Lorentzian function representing the upper atomic level as an energy band of width $\hbar\gamma$, where γ^{-1} is the lifetime of the upper state. Section IV deals with the case where the applied vortex beam is a Laguerre-Gaussian beam. The results are illustrated in Sec. V for the quadrupole atomic transition $6\,^2S_{1/2} \rightarrow 5\,^2D_{5/2}$ in Cs. A summary of our results and brief comments on their significance are given in Sec. VI.

II. QUADRUPOLE RABI FREQUENCY

The quantum system consists of a two-level atom interacting with a single optical vortex beam propagating along the $+z$ axis. The ground and excited states of the two-level atom are $\{|g\rangle, |e\rangle\}$, with level energies $\mathcal{E}_1$ and $\mathcal{E}_2$, respectively, which correspond to a transition frequency $\omega_a = (\mathcal{E}_2 - \mathcal{E}_1)/\hbar$. The interaction Hamiltonian is written as a multipolar series expansion about the center-of-mass coordinate $\mathbf{R}$ as [6,8,10,19]

$$\hat{H}_{\rm int} = \hat{H}_{\rm dp} + \hat{H}_{\rm qp} + \ldots, \tag{1}$$

where the first term $\hat{H}_{\rm dp} = -\hat{\boldsymbol{\mu}} \cdot \hat{\mathbf{E}}(\mathbf{R})$ stands for the electric dipole interaction between the atom and the electric field, $\hat{\boldsymbol{\mu}} = e\mathbf{r}$, with $\mathbf{r}$ the internal position vector, is the electric dipole moment vector, and $\hat{\boldsymbol{E}}(\mathbf{R})$ is the electric field vector. The optical transition in question is taken here to be dipole forbidden but quadrupole allowed, so it is the second (quadrupole) interaction term in Eq. (1) that is of relevance here. We have

$$\hat{H}_{qp} = -\frac{1}{2}\sum_{ij} \hat{Q}_{ij} \nabla_i \hat{E}_j. \tag{2}$$

Here x_i are the components of the internal position vector $\mathbf{r} = (x, y, z)$ and ∇_i are the components of the gradient operator which act only on the spatial coordinates of the transverse electric field vector $\mathbf{E}$ as a function of the center-of-mass position vector variable $\mathbf{R} = (X, Y, Z)$. The quadrupole tensor operator $\hat{Q}_{ij}$ can be written in terms of ladder operators as $\hat{Q}_{ij} = Q_{ij}(\hat{\pi} + \hat{\pi}^\dagger)$, where $Q_{ij} = \langle i|\, \hat{Q}_{ij}\, |j\rangle$ are the quadrupole matrix elements between the two atomic levels, and $\hat{\pi}$ ($\hat{\pi}^\dagger$) are the atomic level lowering (raising) operators.

Without loss of generality, we assume that the electric field is plane polarized along the x direction, so optical spin plays no role here, in which case we have the following form of the quadrupole interaction Hamiltonian:

$$\hat{H}_{\rm qp} = -\frac{1}{2}\sum_i \hat{Q}_{ix} \frac{\partial \hat{E}_x}{\partial R_i}. \tag{3}$$

The quantized electric field can conveniently be written in terms of the center-of-mass position vector in cylindrical polar coordinates $\mathbf{R} = (\rho, \phi, Z)$ as

$$\hat{\mathbf{E}}(\mathbf{R}) = \hat{\mathbf{i}} u_{\{k\}}(\mathbf{R}) \hat{a}_{\{k\}} e^{i\theta_{\{k\}}(\mathbf{R})} + \text{H.c.}, \tag{4}$$

where $u_{\{k\}}(\mathbf{R})$ and $\theta_{\{k\}}(\mathbf{R})$ are, respectively, the amplitude function and the phase function of the LG vortex electric field. Here the subscript $\{k\}$ denotes a group of indices that specify the optical mode in terms of its axial wave vector k, winding number ℓ, and radial number p. The operators $\hat{a}_{\{k\}}$ and $\hat{a}^\dagger_{\{k\}}$ are the annihilation and creation operators of the field mode $\{k\}$. Finally, H.c. stands for Hermitian conjugate. Using this form of the electric field, we obtain the desired expression for the quadrupole interaction Hamiltonian,

$$\hat{H}_{\rm qp} = \hbar\Omega^Q_{\{k\}}(\mathbf{R}) e^{i\theta_{\{k\}}(\mathbf{R})} \hat{a}_{\{k\}} (\hat{\pi}^\dagger + \hat{\pi}) + \text{H.c.}, \tag{5}$$

where $\Omega^Q_{\{k\}}(\mathbf{R})$ is the quadrupole Rabi frequency, which can be written as

$$\Omega^Q_{\{k\}}(\mathbf{R}) = -\frac{1}{2\hbar}\sum_i Q_{ix} u_{\{k\}} \left(\frac{1}{u_{\{k\}}} \frac{\partial u_{\{k\}}}{\partial R_i} + i \frac{\partial \theta_{\{k\}}}{\partial R_i} \right). \tag{6}$$

It is convenient to proceed as we show below by assuming a general LG mode $\text{LG}_{\ell p}$ of winding number ℓ and radial number p. The values of ℓ and p applicable to a given quadrupole transition are decided by application of the selection rules of the specific atomic transition.

III. FERMI GOLDEN RULE FOR ABSORPTION RATE

The vortex field is endowed with an orbital angular momentum $\pm\ell\hbar$ per photon with ℓ positive. Thus, the transition matrix element [20], including only the quadrupole coupling, is given by

$$\mathrm{T}^{\{k\}}_{if} = \langle f|\, \hat{H}_{\rm qp}\, |i\rangle\,, \tag{7}$$

where $|i\rangle$ and $|f\rangle$ are, respectively, the initial and final states of the overall quantum system (atom plus optical vortex). We assume that the system has as an initial state $|i\rangle$ with the atom in its the ground state and there is one vortex photon. The final state $|f\rangle$ consists of the excited state of the atom and there is no field mode. Thus $|i\rangle = |g\{1\}_{\{k\}}\rangle$ and $|f\rangle = |e\{0\}\rangle$:

$$\mathrm{T}^{\{k\}}_{if} = -\frac{1}{2}\sum_{ij} \langle e|\, \hat{Q}_{ij}\, |g\rangle\, \langle\{0\}|\, \frac{\partial \hat{E}_j}{\partial R_i}\, |\{1\}_{\{k\}}\rangle\,. \tag{8}$$

We have taken the electric field to be polarized along the x direction, and using the relations $\langle\{0\}|\, \hat{a}^+_{\{k'\}}\, |\{1\}_{\{k\}}\rangle = 0$ and $\langle\{0\}|\, \hat{a}_{\{k'\}}\, |\{1\}_{\{k\}}\rangle = \delta_{\{k'\}\{k\}}$ we obtain

$$\begin{aligned} \mathrm{T}^{\{k\}}_{if} &= -\frac{1}{2}\sum_i \langle e|\, \hat{Q}_{ix}\, |g\rangle\, \langle\{0\}|\, \frac{\partial \hat{E}_x}{\partial R_i}\, |\{1\}_{\{k\}}\rangle \\ &= \hbar\Omega^Q_{\{k\}}(\mathbf{R}) e^{i\theta_{\{k\}}(\mathbf{R})}, \end{aligned} \tag{9}$$

where $\Omega^Q_{\{k\}}(\mathbf{R})$ is the quadrupole Rabi frequency. The final state of the system in the absorption process consists of a continuous band of energy of width $\hbar\gamma$, where γ is the spontaneous emission rate in free space. In this case the absorption rate is given in the form of Fermi's Golden Rule [21–24] with a density of states

$$\begin{aligned} \Gamma_{if} &= \frac{2\pi}{\hbar^2} \left|\mathrm{T}^{\{k\}}_{if}\right|^2 \mathcal{F}_{\omega_a}(\omega) \\ &= 2\pi \left|\Omega^Q_{\{k\}}(\mathbf{R})\right|^2 \mathcal{F}_{\omega_a}(\omega), \end{aligned} \tag{10}$$

where the density of states is such that $\mathcal{F}_{\omega_a}(\omega)d\omega$ is the number of upper atomic states that fall within the frequency range ω_a to $\omega_a + d\omega$. The density of final states is a function

that peaks at the line center defined by $\hbar\omega_a = \mathcal{E}_2 - \mathcal{E}_1$ and is normalized so that

$$\int_{-\infty}^{\infty} \mathcal{F}_{\omega_a}(\omega)d\omega = 1. \tag{11}$$

The density of states is represented well by a Lorentzian distribution of states with a width (FWHM) coinciding with the spontaneous quadrupole emission rate, thus

$$\mathcal{F}_{\omega_a}(\omega) = \frac{1}{\pi}\frac{\gamma/2}{(\omega-\omega_a)^2 + (\gamma/2)^2}. \tag{12}$$

This function representing the density of states provides a limit to the validity of using Fermi's Golden Rule to evaluate the absorption rate, since this rate is valid only if the frequency width of the upper state $|e\rangle$ is higher than the excitation rate; i.e., the spontaneous emission rate is higher than the Rabi frequency. For high intensities, the Rabi frequency may exceed the spontaneous emission rate, in which case the perturbative approach culminating in the Fermi Golden Rule is no longer valid and the strong-coupling regime is applicable to Rabi oscillations. The maximum value of the density of states as a function of ω is $\frac{2}{\pi\gamma}$, located at $\omega = \omega_a$. Substituting Eq. (12) in Eq. (10) we find for the quadrupole absorption rate

$$\Gamma_{if} = \frac{\gamma}{(\omega-\omega_a)^2 + (\gamma/2)^2}\left|\Omega^Q_{\{k\}}(\mathbf{R})\right|^2. \tag{13}$$

We can now proceed to evaluate the absorption rate when the optical vortex is an LG mode.

IV. ABSORPTION OF A LAGUERRE-GAUSSIAN MODE

In the paraxial regime the quadrupole Rabi frequency associated with the $\mathrm{LG}_{\ell p}$ of frequency ω, which is plane polarized along the x direction, can be written as [6,25–30]

$$\Omega^Q_{k\ell p}(\rho) = \left(u_p^\ell(\rho)/\hbar\right)(U(\mathbf{R})Q_{xx} + V(\mathbf{R})Q_{yx} + ikQ_{zx}), \tag{14}$$

where the functions $U(\mathbf{R})$ and $V(\mathbf{R})$ are

$$U(\mathbf{R}) = \left(\frac{|\ell|X}{\rho^2} - \frac{2X}{w_0^2} - \frac{i\ell Y}{\rho^2} + \frac{1}{L_p^{|\ell|}}\frac{\partial L_p^{|\ell|}}{\partial X}\right), \tag{15}$$

$$V(\mathbf{R}) = \left(\frac{|\ell|Y}{\rho^2} - \frac{2Y}{w_0^2} + \frac{i\ell X}{\rho^2} + \frac{1}{L_p^{|\ell|}}\frac{\partial L_p^{|\ell|}}{\partial Y}\right), \tag{16}$$

and

$$u_{\{k\}}(\rho) = u_{k\ell p}(\rho) = E_{k00}f_{\ell,p}(\rho), \tag{17}$$

with

$$f_{\ell,p}(\rho) = \sqrt{\frac{p!}{(|\ell|+p)!}}\left(\frac{\rho\sqrt{2}}{w_0}\right)^{|\ell|} L_p^{|\ell|}\left(\frac{2\rho^2}{w_0^2}\right)e^{-\rho^2/w_0^2}, \tag{18}$$

where $L_p^{|\ell|}$ is the associated Laguerre polynomial and w_0 is the radius at the beam waist (at $Z = 0$). The overall factor E_{k00} is the constant amplitude of the corresponding plane electromagnetic wave. The phase function of the LG mode in the paraxial regime is as follows:

$$\theta_{klp}(\rho, Z, t) \approx kZ + l\phi - \omega t. \tag{19}$$

Substituting in Eq. (13) we have the quadrupole absorption rate for an atom interacting with the $\mathrm{LG}_{\ell,p}$ light mode that is polarized along the x direction and the atom is characterized by the three quadrupole matrix elements Q_{xx}, Q_{xy}, and Q_{xz}:

$$\Gamma_{if} = \frac{\gamma}{(\omega-\omega_a)^2 + (\gamma/2)^2}|(U(\mathbf{R})Q_{xx} + V(\mathbf{R})Q_{yx} + ikQ_{zx})|^2 \times \left|u_p^\ell(\rho)/\hbar\right|^2. \tag{20}$$

So far the treatment has been general and Eq. (20) is the main result of this paper. This result applies to any atom with a dipole-forbidden but quadrupole-allowed transition which is at near-resonance with a linearly polarized Laguerre-Gaussian light mode $\mathrm{LG}_{\ell,p}$. The main requirement is that the interaction must conform with the OAM selection rules involving the quantum number m between the ground and the excited atomic states $|g\rangle$ and $|e\rangle$, and we have for a quadrupole transition

$$\mathbf{\Delta}m = 0, \pm 1, \pm 2. \tag{21}$$

The requirement for OAM conservation then means that the optical vortex absorption process in a quadrupole transition can only occur for optical vortices with winding numbers $\ell = 0, +1, +2$. The case $\ell = 0$ is possible, but then no transfer of OAM occurs in the absorption process, while each of the cases $\ell = 1$ and $\ell = 2$ is accompanied by a transfer of OAM of magnitudes $\hbar$ and $2\hbar$, respectively. The details will depend on the specific atom and its specific quadrupole transition. Note that although the radial quantum number p is important for the amplitude distribution function of the $\mathrm{LG}_{\ell,p}$ mode, the magnitude of the OAM transferred is determined solely by the value of the winding number $\ell \leqslant 2$.

In order to illustrate the main result with practical examples, we focus on a case that has recently been discussed [6,8,10,19], namely, an LG mode of winding number $\ell = 0, 1, 2$ and radial number p. In the simplest case, where the mode is a doughnut mode $p = 0$, we find that the last terms involving the derivatives in $U(\mathbf{R})$ and $V(\mathbf{R})$ given by Eqs. (15) and (16) vanish, as $L_0^{|\ell|}$ are constants for all ℓ. However, the case where $p \neq 0$ is also of interest since the value of p is important for the intensity distribution. A specific atomic transition we consider to illustrate the results is that of the neutral cesium atom, namely, the $6\,^2S_{1/2} \rightarrow 5\,^2D_{5/2}$ transition. However, in order to proceed with evaluations, we need the values of the quadrupole matrix elements Q_{xx}, Q_{xy}, and Q_{xz} applicable in the transition, depending on the OAM selection rules.

V. VORTEX ABSORPTION IN CESIUM

The quadrupole matrix elements Q_{xx}, Q_{xy}, and Q_{xz} can be discussed with reference to the normalized hydrogenlike wave function ψ_{nLm} [31,32],

$$\psi_{nLm}(r,\theta,\phi) = \left\{\left(\frac{2Z_a}{na_\mu}\right)^3\frac{(n-L-1)!}{2n(n+L)!}\right\}^{1/2} \times e^{-\rho(r)/2}\rho(r)^l \mathrm{L}^{2L+1}_{n-L-1}(\rho(r))Y_L^m(\theta,\phi), \tag{22}$$

where $a_\mu = \frac{4\pi\epsilon_0\hbar^2}{\mu e^2} = a_0\frac{m_e}{\mu}$ is the reduced Bohr radius, $\mathrm{L}^{2L+1}_{n-L-1}(\rho(r))$ are the associated Laguerre polynomials, and $\rho(r) = \frac{2Z_a r}{na_\mu}$. The valence electron of the Cs atom sees an effective nuclear charge of $Z_a = 8.56$ [9,31–35]. Using Eq. (22),

we have for the electric quadrupole matrix element

$$Q_{\alpha\beta} = e\,\langle\psi_f|\,x_\alpha x_\beta\,|\psi_i\rangle\,, \tag{23}$$

where $x_\alpha = (x, y, z)$. Straightforward evaluations yield the following:

(i) for the case $\Delta m = 0$, we find that $Q_{xx} = \frac{70.2}{Z_a^2} e a_\mu^2$ and $Q_{xy} = Q_{xz} = 0$;

(ii) for the case $\Delta m = \pm 1$, we have $Q_{zx} = i\frac{86}{Z_a^2} e a_\mu^2$ and $Q_{xx} = Q_{yx} = 0$; and

(iii) for $\Delta m = \pm 2$, we have $Q_{xx} = \pm i Q_{yx} = \frac{86}{Z_a^2} e a_\mu^2$ and $Q_{zx} = 0$.

We consider a quadrupole transition with the selection rule $\Delta m = 1$ applicable to the $(6\,^2S_{1/2} \to 5\,^2D_{5/2})$ quadrupole transition in Cs. In this case, the quadrupole moments are $Q_{xx} = Q_{xy} = 0$ and $Q_{xz} \neq 0$ and the Rabi frequency, Eq. (14), is as follows:

$$\Omega^Q_{k\ell 0}(\rho) = ikQ_{zx}\big(u_0^{|\ell|}(\rho)/\hbar\big). \tag{24}$$

The absorption rate is then given by

$$\Gamma_{if} = \frac{\gamma}{(\omega-\omega_a)^2 + (\gamma/2)^2}\left|\Omega^Q_{\{k\}}(\mathbf{R})\right|^2 \tag{25}$$

$$= \frac{2\pi w_0^2}{c^2}|\Omega_0|^2 F(\omega,\omega_a)|f_{\ell,0}(\rho)|^2, \tag{26}$$

where $f_{\ell,0}$ is given by Eq. (18) with $p = 0$, Ω_0 is a scaling factor for the Rabi frequency,

$$\Omega_0 = \frac{1}{\hbar}\frac{E_{k00}Q_{zx}}{w_0}, \tag{27}$$

and the modified density function $F(\omega, \omega_a)$ is given by

$$F(\omega,\omega_a) = \frac{\gamma/2}{\pi}\frac{\omega^2}{(\omega-\omega_a)^2 + (\gamma/2)^2}, \tag{28}$$

where the maximum value of this function is $\frac{1}{\pi}(\frac{\omega_a^2}{\gamma/2} + \gamma/2)$, located at $\omega = \omega_a + \frac{(\gamma/2)^2}{\omega_a}$. It is clear that the dependence of the Rabi frequency on the light frequency affects the transition rate. However, the maximum of the function $F_{\omega_a}(\omega)$ is greater than that of the density of states and its position is shifted away from ω_a. In Fig. 1, we present the shape of the normalized density of state $F_{\omega_a}(\omega)$ in terms of the ratio ω/ω_a for different values of γ/ω_a.

Typical parameters in this case are [36] $\lambda = 685$ nm, $Q_{zx} \simeq 10ea_0^2$, and the spontaneous decay rate is $\Gamma_S = 3.34 \times 10^7\ \mathrm{s}^{-1}$ [37,38]. The beam parameters are chosen such that the beam waist $w_0 = \lambda d$, where d is a real number, and the intensity $I = \epsilon_0 c E_{k00}^2/2$. Introducing the dimensionless magnitude of the intensity $\mathcal{I} = I/I_0$, where $I_0 = 1\ \mathrm{W\,m^{-2}}$, the scaling factor of the Rabi frequency can be written as

$$\Omega_0 = \frac{1}{\hbar}\left(\frac{2I}{\epsilon_0 c}\right)^{1/2}\frac{Q_{zx}}{w_0} = 5.14\times 10^{-5}\frac{(\mathcal{I})^{1/2}}{d}\Gamma_S. \tag{29}$$

We must make an appropriate choice of the beam waist and the magnitude of the intensity of the field and ensure that $\Omega_0 \ll \Gamma_S$, which is the condition for the validity of the Fermi Golden Rule.

It is clear that for a weak intensity, we can obtain an absorption rate Γ_{if} lower than the spontaneous transition Γ_S. On the other hand, the Lorentzian density of states is chosen

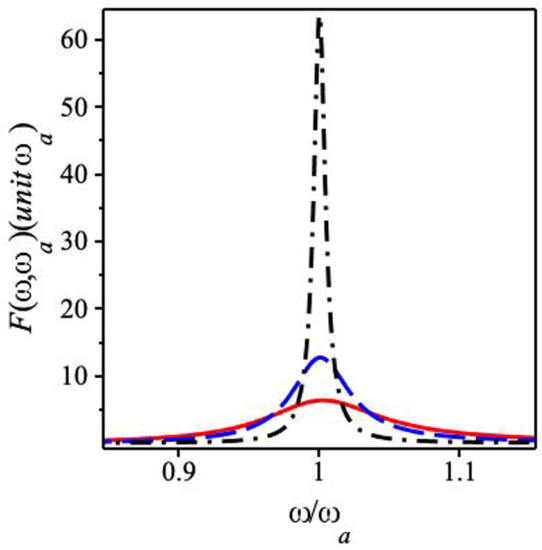

FIG. 1. The normalized modified density function $F(\omega, \omega_a)$ (unit, ω_a) as a function of the ratio ω/ω_a. The solid red curve represents $\gamma/\omega_a = 0.1$; the dashed blue curve, $\gamma/\omega_a = 0.05$; and the dash-dotted black curve, $\gamma/\omega_a = 0.01$. Note the tendency of $F(\omega, \omega_a)$ to become a function proportional to a delta function as the value of γ/ω_a decreases.

with a width given by the spontaneous emission rate $\gamma = \Gamma_S$, where $\Gamma_S \ll \omega_a$; thus the transition rate can be written as

$$\Gamma_{if} = 9.64\times 10^{-23}\mathcal{I}\left(\frac{\Gamma_S}{2\pi}\right)\frac{\omega^2}{(\omega-\omega_a)^2 + (\Gamma_S/2)^2}|f_{\ell,0}(\rho)|^2. \tag{30}$$

We assume a moderate laser intensity, $I = 40 \times 10^4\ \mathrm{W\,m^{-2}}$ [36], and substituting for the relevant parameter values we have for the absorption rate at $\omega = \omega_a$

$$\Gamma_{if} = 1.67\times 10^{-1}\Gamma_S|f_{\ell,0}(\rho)|^2, \tag{31}$$

which suggests that the absorption rate is much lower than the spontaneous rate, depending on the relative position of the atom. In Fig. 2, we present the variation of the absorption rate Γ_{if}/Γ_S as a function of the radial position of the atom ρ/λ for different values of the beam waist, $w_0/\lambda = 2$, 5, and 10. It is clear that the maximum of the rate shifts away from the origin with increasing beam waist w_0, but the value of the maximum is the same and is independent of w_0. This observation can be confirmed analytically as follows. Equation (26) shows that Γ_{if} is proportional to w_0^2 and to Ω_0^2, but Ω_0^2 is inversely proportional to w_0^2 as shown by Eq. (28). Thus, apart from constant factors, the relevant dependence of Γ_{if} on w_0 resides essentially in the function $|f_{\ell,p}(\rho)|^2$ given by Eq. (18), which, apart from constant factors for $\ell = 1$ and $p = 0$, is as follows:

$$|f_{1,0}(\rho/w_0)|^2 = \frac{2\rho^2}{w_0^2}e^{-2\rho^2/w_0^2}. \tag{32}$$

It is easy to show that the maximum of this function occurs at the radial position $\rho = w_0/\sqrt{2}$ and the value of the maximum rate is $1/e$, which is independent of w_o, as shown in Fig. 2(a).

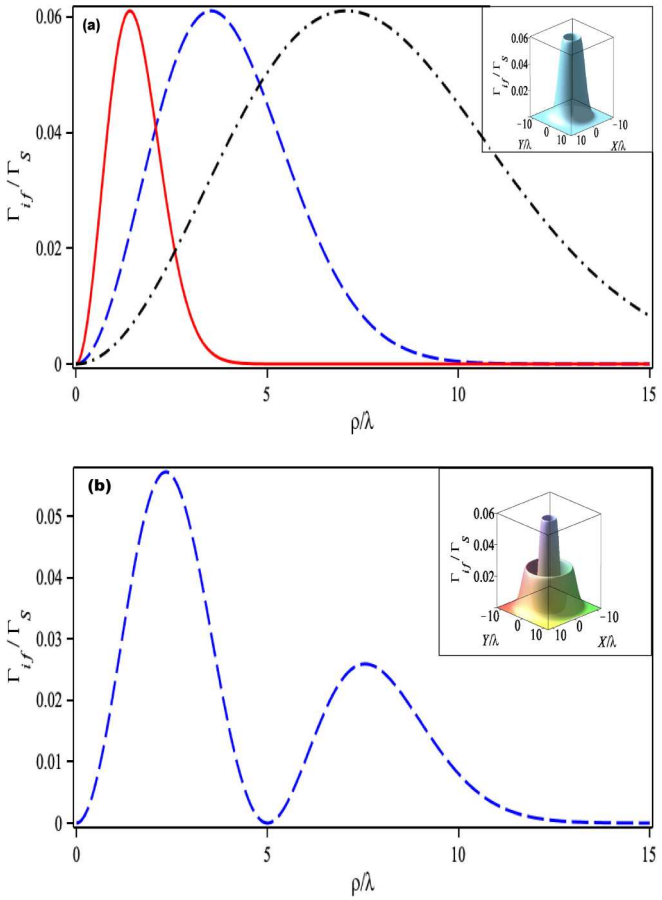

FIG. 2. The variation with radial position of the quadrupole absorption rate Γ_{if}/Γ_S for $\Delta m = +1$, (a) $\ell = 1$, $p = 0$, and (b) $\ell = 1$, $p = 1$, and an atom in a Laguerre-Gaussian mode $\mathrm{LG}_{\ell,p}$. The solid red line represents the case $w_0/\lambda = 2$; the dashed blue line, $w_0/\lambda = 5$; and the dash-dotted line, $w_0/\lambda = 10$. Insets: The cylindrically symmetric quadrupole rate for the case $w_0/\lambda = 5$. Note that in (a) the maxima of the absorption rates for plots with different w_0's have the same value independent of w_0. See the text for the derivation confirming this observation.

However, for the case $\ell = 1$ and $p = 1$, we have

$$|f_{1,1}(\rho/w_0)|^2 = 2\frac{\rho^2}{w_0^2}\left(1 - \frac{\rho^2}{w_0^2}\right)e^{-2\rho^2/w_0^2}. \qquad (33)$$

There are two maxima of this function in this case as in Fig. 2(b). These occur at the radial positions $\rho = \frac{w_0}{2}\sqrt{5 \pm \sqrt{17}}$ and the maximum rates at these positions are, once more, independent of w_o.

VI. CONCLUSION

This paper has focused on the interaction of atoms with light endowed with orbital angular momentum, where our main aim was to evaluate the rate of transfer of OAM from the light to the atoms in a dipole-forbidden but quadrupole-allowed transition. Our work follows two significant developments. The first development concerns the latest experimental confirmation by Giammanco *et al.* [17] following earlier experiments [14–16] that OAM cannot be transferred to the internal (electronic-type) degrees of freedom of the atom in an electric dipole transition, though a transfer to the internal degrees of freedom of atoms was predicted theoretically to occur in the normally much weaker quadrupole transition [6]. The second development concerns the very recent considerable advances in the ability to carry out delicate measurements in experiments targeted specifically at quadrupole transitions involving light carrying OAM [37,39,40]. However, although such experiments have demonstrated the involvement of optical vortices with atoms in quadrupole transitions, the rate of OAM transfer in a quadrupole transition has not, as far as we know, been evaluated, so our task in this work involved setting up the theory of optical vortex photon absorption by an atom in a quadrupole-allowed transition. Our theory is general and applies to any atom with a quadrupole transition, as, for example, in Na and Rb atoms, both of which have been the subject of investigations in connection with optical vortex interaction with atoms. However, we proceeded to apply the theory to the well-known case of the Cs $6\,^2S_{1/2} \rightarrow 5\,^2D_{5/2}$ quadrupole transition, which conforms with the requirements for OAM conservation consistent with the rules $\Delta m = 0, \pm 1, \pm 2$.

Absorption for the case $\Delta m = +1$ required an optical vortex in the form of a Laguerre-Gaussian mode with $\ell = 1$ and we have considered two modes, one with $p = 0$ and the second with $p = 1$. We have found that the absorption rate as a function of the radial position mirrors the intensity distribution of the beam. For the doughnut mode the rates peak at the atomic location of $\rho = w_0\sqrt{\ell/2}$, while for the case $p = 1$ the maxima are located at $\rho/w_0 = \frac{1}{2}\sqrt{2\ell + 3 \pm \sqrt{8\ell + 9}}$. Note that the maximum rate is a constant, i.e., independent of w_0, and merely shifts its position as w_0 increases.

The maximum magnitude of the absorption rate in the example we have considered for Cs quadrupole transitions, assuming an intensity of about $10^5\,\mathrm{W\,m^{-2}}$, is of the order of 6% of the quadrupole spontaneous emission rate. Such a magnitude should not be beyond the capability of current spectroscopic techniques.

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