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Angular momentum transfer in interaction of Laguerre-Gaussian beams with atoms and molecules

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The exchange of orbital angular momentum between a Laguerre-Gaussian beam of light and the center-of-mass motion of an atom or molecule is well known. We show that the orbital angular momentum of light can also be transferred to the internal electronic or rotational motion of an atom or a molecule provided the internal and center-of-mass motions are coupled. However, this transfer does not happen directly to the internal motion, but via center-of-mass motion. If atoms or molecules are cooled down to the recoil limit, then an exchange of angular momentum between the quantized center-of-mass motion and the internal motion is possible during the interaction of cold atoms or molecules with the Laguerre-Gaussian beam. The orientation of the exchanged angular momentum is determined by the sign of the winding number of the Laguerre-Gaussian beam. We present selective results of numerical calculations for the quadrupole transition rates in the interaction of the Laguerre-Gaussian beam with an atomic Bose-Einstein condensate to illustrate the underlying mechanism of light orbital angular momentum transfer. We discuss how the alignment of diatomic molecules will facilitate exploration of the effects of light orbital angular momentum on the electronic motion of molecules.

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I. INTRODUCTION

Two decades after the pioneering work of Allen and coworkers [1] showing that Laguerre-Gaussian (LG) beams carry well-defined orbital angular momentum (OAM), the role of this OAM in interactions of such beams with an atom or a molecule remains an open question. OAM of light usually interacts with the external center-of-mass (c.m.) motion of an atom [2,3]. In contrast, light polarization, which is the spin angular momentum of light, can interact with the internal electronic motion of an atom. Many researchers have predicted that the field OAM can be transferred to the internal motion of an atom [4–8] or a molecule [9–11] in electronic dipole or quadrupole transitions, while some works [12,13] have shown that the field OAM does not interact with molecular chirality. Applications have been proposed based on direct coupling of field OAM to the internal motion [4,7,8,11,14–17] or to the c.m. motion only [18–20]. The transfer of field OAM to the c.m. motion of optically trapped particles (optical spanner effect) [21–23] and Bose-Einstein condensates (BEC) [24,25] is well known. But the experiments so far seem to contradict direct coupling of field OAM with internal motion [26,27]. It is therefore important to understand how light OAM takes part in a light-atom or light-molecule interaction. The OAM of light is associated with the spatial inhomogeneity of field over the beam cross section. The question we address here is, Can an electron in an atom or a molecule feel the spatial variation of the field during its orbital motion?

This paper is organized as follows. In Sec. II, we develop the theory corresponding to the method of OAM exchange in the interaction of the LG beam with an atom or a molecule separately. Section III presents numerical calculations of transition rates in the interaction of atomic BEC with an LG beam as an example of our theory. Finally, in Sec. IV, we make some concluding remarks.

II. THEORY

We consider an LG beam without any off-axis node propagating along the z axis of the laboratory frame interacting with a cold atom or a cold molecule whose c.m. wave function has an extension comparable to the wavelength of the light but smaller than the waist of the beam. The atom or molecule experiences a local field of the type [6,10,11]

$$\mathbf{E}(\mathbf{r}',t) = \frac{\mathbf{E}_0}{\sqrt{|l|!}}\left(\frac{r'_\perp}{w_0}\right)^{|l|}\exp(il\phi')\exp[i(kz'-\omega t)], \tag{1}$$

where $r'_\perp$ is the projection of $\mathbf{r}'$ on the xy plane, l and w_0 are the winding number and the waist of the beam, respectively, and ϕ' is the azimuth.

(a) The atom-radiation interaction. We consider the simplest atomic system composed of a nucleus of positive charge (mass) $+e(m_n)$ and an electron of negative charge $-e(m_e)$. For simplicity, the spin of the particles is ignored. The center-of-mass coordinate of the atomic system is $\mathbf{R}_{\text{c.m.}} = (m_e\mathbf{r}_e + m_n\mathbf{r}_n)/m_t$, with $m_t = m_e + m_n$ being the total mass and $\mathbf{r}_e$ and $\mathbf{r}_n$ being the coordinates of the electron and nucleus, respectively. The Hamiltonian of the atom-field system is $H = H_0 + H_{\text{I,atom}}$, where H_0 is the unperturbed atomic Hamiltonian and

$$H_{\text{I,atom}} = -\int d\mathbf{r}'\mathcal{P}(\mathbf{r}')\cdot\mathbf{E}(\mathbf{r}',t) + \text{H.c.} \tag{2}$$

is the interaction Hamiltonian derived in the Power-Zineau-Wooley (PZW) scheme [9,10,28]. $\mathcal{P}(\mathbf{r}')$ is the electric polarization given by

$$\mathcal{P}(\mathbf{r}') = -e\frac{m_n}{m_t}\mathbf{r}\int_0^1 d\lambda\,\delta\left(\mathbf{r}' - \mathbf{R}_{\text{c.m.}} - \lambda\frac{m_n}{m_t}\mathbf{r}\right), \tag{3}$$

where the relative coordinate (internal) $\mathbf{r} = \mathbf{r}_e - \mathbf{r}_n$.

The diameter of the region of the LG beam of Eq. (1) typically ranges between 10^{-4} and 10^{-5} m [24,29–33], while the dimension of an electron orbital in an atom is of the order of a few angstroms. This means $|\mathbf{r}| \ll |\mathbf{R}_{\text{c.m.}}|$, and we therefore

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use Taylor's expansion

$$\mathbf{E}\left(\mathbf{R}_{\text{c.m.}}+\lambda\frac{m_n}{m_t}\mathbf{r}\right)=\mathbf{E}(\mathbf{R}_{\text{c.m.}})+\frac{\mathbf{E}_0}{\sqrt{|l|!}}\left(\lambda\frac{m_n}{m_t}\right)\left(\frac{1}{w_0}\right)^{|l|}\times r\{\hat{\mathbf{r}}\cdot\vec{\nabla}(r_\perp'^{|l|}e^{il\phi'}e^{ikz'})\}_{\mathbf{R}_{\text{c.m.}}}+\cdots, \tag{4}$$

where

$$\begin{aligned} r\{\hat{\mathbf{r}}\cdot\vec{\nabla}(r_\perp'^{\,|l|}e^{il\phi'}e^{ikz'})\}_{\mathbf{R}_{\text{c.m.}}} &= R_{\text{c.m.}\perp}^{|l|-1}e^{il\Phi_{\text{c.m.}}}e^{ikZ_{\text{c.m.}}}\tfrac{1}{2}[e^{i\phi}e^{-i\Phi_{\text{c.m.}}}(|l|+l)\\ &\quad +e^{-i\phi}e^{i\Phi_{\text{c.m.}}}(|l|-l)]r\sin\theta\\ &\quad +(ik)R_{\text{c.m.}\perp}^{|l|}e^{il\Phi_{\text{c.m.}}}e^{ikZ_{\text{c.m.}}}r\cos\theta. \end{aligned} \tag{5}$$

Substituting Eqs. (3)–(5) into Eq. (2), $H_{\text{I,atom}}$ can be separated into the dipole and quadrupole parts as given by

$$H_{\text{I,atom}}^{d}=\sqrt{\frac{4\pi}{3|l|!}}e\left(\frac{m_n}{m_t}\right)r\sum_{\sigma=0,\pm1}\epsilon_\sigma Y_1^\sigma(\hat{\mathbf{r}})\left(\frac{R_{\text{c.m.}\perp}}{w_0}\right)^{|l|}\times e^{il\Phi_{\text{c.m.}}}e^{ikZ_{\text{c.m.}}}+\text{H.c.}, \tag{6}$$

$$\begin{aligned} H_{\text{I,atom}}^{q}=\frac{1}{2}\sqrt{\frac{4\pi}{3|l|!}}e\left(\frac{m_n}{m_t}\right)^2\left(\frac{1}{w_0}\right)^{|l|}r^2\sum_{\sigma=0,\pm1}\epsilon_\sigma Y_1^\sigma(\hat{\mathbf{r}})\\ \times\Big\{R_{\text{c.m.}\perp}^{|l|-1}e^{il\Phi_{\text{c.m.}}}e^{ikZ_{\text{c.m.}}}\frac{1}{2}[e^{i\phi}e^{-i\Phi_{\text{c.m.}}}(|l|+l)\\ +e^{-i\phi}e^{i\Phi_{\text{c.m.}}}(|l|-l)]\sin\theta\\ +(ik)R_\perp^{|l|}e^{il\Phi_{\text{c.m.}}}e^{ikZ_{\text{c.m.}}}\cos\theta\Big\}+\text{H.c.} \end{aligned} \tag{7}$$

The dot products of the type $\mathbf{r}'\cdot\mathbf{E}_0$ are replaced by $r'\sqrt{4\pi/3}\sum_{\sigma=0,\pm1}\epsilon_\sigma Y_1^\sigma(\theta',\phi')$, with $\epsilon_{\pm1}=(E_x\pm iE_y)/\sqrt{2}$ and $\epsilon_0=E_z$. In paraxial approximation, the E_z component is negligible. Equation (6) shows that within the electric dipole approximation the polarization of the field interacts with the electronic motion, and the field OAM interacts only with the external c.m. motion, as also demonstrated by several authors [2,4,6,9]. The first term in Eq. (7) implies that in the electric quadrupole transition, the field OAM is coupled to the c.m. motion only and the extra unit of angular momentum in the electronic motion results from the quantized c.m. motion of the atom as in Ref. [2]. The invariance of the interaction Hamiltonian around the beam axis imposes the conservation of total angular momentum of the field-plus-atom system, while the gradient of the field along the radial direction couples the quantized c.m. and electronic motions. This coupling of c.m. and electronic motions is the main novel feature of this interaction. Our calculation shows that either of the terms $(|l|+l)$ or $(|l|-l)$ is nonzero depending on the sign of l. The quadrupole transition matrix element is given by $\mathcal{M}_{i\to f}^{q}=\langle\Upsilon_f|H_{\text{I,atom}}^{q}|\Upsilon_i\rangle$, where Υ denotes an unperturbed atomic state, i.e., eigenstate of H_0. We assume $\Upsilon(\mathbf{R}_{\text{c.m.}},\mathbf{r})=\Psi_{\text{c.m.}}(\mathbf{R}_{\text{c.m.}})\psi(\mathbf{r})$, where the c.m. wave function $\Psi_{\text{c.m.}}(\mathbf{R}_{\text{c.m.}})$ depends on the external potential that traps the atom and the internal electronic wave function $\psi(\mathbf{r})$ can be considered to be a highly correlated coupled-cluster orbital [34].

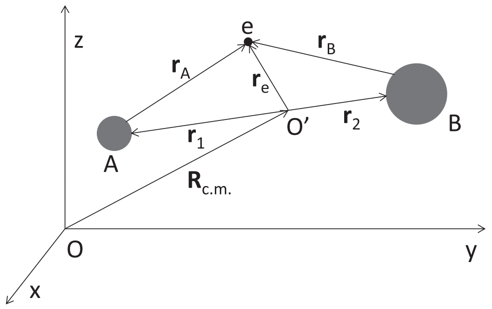

FIG. 1. Diatomic molecule in the laboratory coordinate system. The z axis is along the direction of propagation of the LG beam.

$$\begin{aligned} \mathcal{M}_{i\to f}^{q}=\frac{1}{2}\sqrt{\frac{4\pi}{3|l|!}}e\left(\frac{m_n}{m_t}\right)^2\sum_{\sigma=0,\pm1}\epsilon_\sigma\Big\{|l|\left(\frac{w_{\text{c.m.}}}{w_0}\right)^{|l|-1}\\ \times\left(\frac{w_e}{w_0}\right)\langle\psi_f|\frac{r^2}{w_e}Y_1^\sigma(\hat{\mathbf{r}})\sin\theta e^{\text{sgn}(l)i\phi}|\psi_i\rangle\mathcal{M}_{\text{c.m.}}^{\text{sgn}(l)(|l|-1)}\\ +(ik)\left(\frac{w_{\text{c.m.}}}{w_0}\right)^{|l|}\langle\psi_f|r^2Y_1^\sigma(\hat{\mathbf{r}})\cos\theta|\psi_i\rangle\mathcal{M}_{\text{c.m.}}^{l}\Big\}, \end{aligned} \tag{8}$$

where $w_{\text{c.m.}}$ and w_e stand for the average spatial width of $\Psi_{\text{c.m.}}(\mathbf{R}_{\text{c.m.}})$ and $\psi(\mathbf{r})$, respectively, and $\mathcal{M}_{\text{c.m.}}^{l'}=\langle\Psi_{\text{c.m.}f}|(\frac{R_{\text{c.m.}\perp}}{w_{\text{c.m.}}})^{|l'|}e^{il'\Phi_{\text{c.m.}}}e^{ikZ_{\text{c.m.}}}|\Psi_{\text{c.m.}i}\rangle$. From Eq. (8) we deduce the selection rule for magnetic quantum number $\Delta m=0$, ±1, ±2. The extra unit of angular momentum transferred to the electron comes from the quantized c.m. motion of the atom. The orientation of this angular momentum is the same as that of the field OAM. We have assumed that $w_{\text{c.m.}}\gg w_e$. Hence, the transition probabilities become insignificant for higher-order multipole transitions unless the intensity of beam is very high.

(b) The molecule-radiation interaction. We consider a diatomic molecular ion, e.g., H_2^+ or HD^+ for simplicity, comprising three particles: two nuclei of mass (charge) m_1 $(+e)$ and m_2 $(+e)$ and one electron m_e $(-e)$, as schematically shown in Fig. 1. A and B are two nuclei, and O' is the center of mass of the molecule. $\mathbf{r}_1$,$\mathbf{r}_2$ are the positions of nuclei A and B with respect to the c.m., respectively. $\mathbf{r}_e$ is the position of the electron in the c.m. coordinate system. A molecular axis is defined as the position of A with respect to B, i.e., $\overrightarrow{BA}=\mathbf{R}$. The nuclei oscillations q are written explicitly as $\mathbf{R}=\bar{\mathbf{R}}+\mathbf{v}q$, where $\bar{\mathbf{R}}$ is the equilibrium position of A with respect to B and $\mathbf{v}$ is a constant vector with the same direction as that of $\mathbf{R}$ and a magnitude given by the properties of the molecule. $\mathbf{r}_A$ and $\mathbf{r}_B$ are positions of the electron relative to nuclei A and B, respectively. Hence, $\mathbf{r}_e=\mathbf{r}_1+\mathbf{r}_A$, $\mathbf{r}_e=\mathbf{r}_2+\mathbf{r}_B$, and $\mathbf{r}_e=\frac{1}{2}\{r\hat{\mathbf{R}}+(\mathbf{r}_A+\mathbf{r}_B)\}$, where $r=|\mathbf{r}_1|-|\mathbf{r}_2|=\frac{R(m_2-m_1)}{m_2+m_1}$ and $\hat{\mathbf{R}}$ is a unit vector along $\mathbf{R}$. The unperturbed quantum state of the diatomic molecule is described by $\Psi=\psi_e(\mathbf{r}_A,\mathbf{r}_B)\psi_v(q)\psi_r(\hat{\mathbf{R}})\Psi_{\text{c.m.}}(\mathbf{R}_{\text{c.m.}})$, where $\psi_e(\mathbf{r}_A,\mathbf{r}_B)$ stands for the electronic state, $\psi_v(q)$ stands for the vibrational wave function, $\psi_r(\hat{\mathbf{R}})$ stands for the rotational wave function, and $\Psi_{\text{c.m.}}(\mathbf{R}_{\text{c.m.}})$ stands for the c.m. wave function. The interaction Hamiltonian is derived in the PZW scheme as in the previous section. Again, we assume that the molecular dimension is very small compared to the beam

waist ($|\mathbf{r}_e| \ll |\mathbf{R}_{\text{c.m.}}|$) and use Taylor's expansion about the c.m. position,

$$\mathbf{E}(\mathbf{R}_{\text{c.m.}} + \lambda\mathbf{r}_e) = \mathbf{E}(\mathbf{R}_{\text{c.m.}}) + \frac{\mathbf{E}_0}{\sqrt{|l|!}}\lambda\left(\frac{1}{w_0}\right)^{|l|} \times r_e\{\hat{\mathbf{r}}_e \cdot \vec{\nabla}({r'_\perp}^{|l|} e^{il\phi'} e^{ikz'})\}_{\mathbf{R}_{\text{c.m.}}} + \cdots, \quad (9)$$

where

$$r_e\{\hat{\mathbf{r}}_e \cdot \vec{\nabla}({r'_\perp}^{|l|} e^{il\phi'} e^{ikz'})\}_{\mathbf{R}_{\text{c.m.}}} = -2l R_{\text{c.m.}\perp}^{|l|-1} e^{\text{sgn}(l)i(|l|-1)\Phi_{\text{c.m.}}} e^{ikZ_{\text{c.m.}}} \mathcal{R}_1^{\text{sgn}(l)1}(\mathbf{r}_e) + (ik) R_{\text{c.m.}\perp}^{|l|} e^{il\Phi_{\text{c.m.}}} e^{ikZ_{\text{c.m.}}} \mathcal{R}_1^0(\mathbf{r}_e) \quad (10)$$

and $\mathcal{R}_l^m(\mathbf{r}')$ is the regular solid spherical harmonic function given as $\mathcal{R}_l^m(\mathbf{r}') = \mathcal{N}_{l,m}(r')^l Y_l^m(\theta',\phi')$ [6,10]. The normalization constant is $\mathcal{N}_{l,m} = \sqrt{4\pi/(2l+1)(l+m)!(l-m)!}$. Using the expression of $\mathbf{r}_e$ and the translation property of solid spherical harmonics [10,11], Eq. (10) can be expressed as

$$\mathbf{r}_e \cdot \{\vec{\nabla}({r'_\perp}^{|l|} e^{il\phi'} e^{ikz'})\}_{\mathbf{R}_{\text{c.m.}}} = -l R_{\text{c.m.}\perp}^{|l|-1} e^{\text{sgn}(l)i(|l|-1)\Phi_{\text{c.m.}}} e^{ikZ_{\text{c.m.}}} \times \left[\mathcal{R}_1^{\text{sgn}(l)1}(r\hat{\mathbf{R}}) + \mathcal{R}_1^{\text{sgn}(l)1}(\mathbf{r}_A + \mathbf{r}_B)\right] + (ik)(1/2) R_{\text{c.m.}\perp}^{|l|} e^{il\Phi_{\text{c.m.}}} e^{ikZ_{\text{c.m.}}} \times \left[\mathcal{R}_1^0(r\hat{\mathbf{R}}) + \mathcal{R}_1^0(\mathbf{r}_A + \mathbf{r}_B)\right]. \quad (11)$$

The second term of Eq. (9) shows that one unit of angular momentum is transferred from the external c.m. motion to the internal rotational and electronic motion of the molecule. If we retain only the first two terms in Eq. (9) and carry out the integration over λ, we get the dipole interaction Hamiltonian as given by

$$H_{\text{I,mol}}^d = \frac{e}{2}\sqrt{\frac{4\pi}{3|l|!}}\left(\frac{1}{w_0}\right)^{|l|} \sum_{\sigma=0,\pm1} \epsilon_\sigma \Bigg\{ R_{\text{c.m.}\perp}^{|l|} e^{il\Phi_{\text{c.m.}}} e^{ikZ_{\text{c.m.}}} \Bigg[\left[r_A Y_1^\sigma(\hat{\mathbf{r}}_A) + r_B Y_1^\sigma(\hat{\mathbf{r}}_B)\right]\left(1 + \frac{ik}{4}\mathcal{R}_1^0(r\hat{\mathbf{R}})\right) + \frac{ik}{4} r Y_1^\sigma(\hat{\mathbf{R}})\mathcal{R}_1^0(\mathbf{r}_A + \mathbf{r}_B)\Bigg]$$
$$- \frac{l}{2} R_{\text{c.m.}\perp}^{|l|-1} e^{\text{sgn}(l)i(|l|-1)\Phi_{\text{c.m.}}} e^{ikZ_{\text{c.m.}}} \left[r Y_1^\sigma(\hat{\mathbf{R}})\mathcal{R}_1^{\text{sgn}(l)1}(\mathbf{r}_A + \mathbf{r}_B) + \left[r_A Y_1^\sigma(\hat{\mathbf{r}}_A) + r_B Y_1^\sigma(\hat{\mathbf{r}}_B)\right]\mathcal{R}_1^{\text{sgn}(l)1}(r\hat{\mathbf{R}})\right]\Bigg\} + \text{H.c.} \quad (12)$$

The transition matrix element of the dipole interaction Hamiltonian (12) is

$$\mathcal{M}_{i\to f} = \langle\Psi_f|H_{\text{I, mol}}^d|\Psi_i\rangle$$
$$= \frac{e}{2}\sqrt{\frac{4\pi}{3|l|!}} \sum_{\sigma=0,\pm1} \epsilon_\sigma \Bigg\{ \left(\frac{w_{\text{c.m.}}}{w_0}\right)^{|l|} \Bigg\{ \langle\psi_{ef}|\left[r_A Y_1^\sigma(\hat{\mathbf{r}}_A) + r_B Y_1^\sigma(\hat{\mathbf{r}}_B)\right]|\psi_{ei}\rangle \left[\mathcal{M}_{r0}^0\mathcal{M}_v^0 + \frac{ik}{4}\sqrt{\frac{4\pi}{3}}\left(\frac{m_2 - m_1}{m_2 + m_1}\right)\bar{R}\mathcal{M}_{r1}^0\mathcal{M}_v^1\right]$$
$$+ \left(\frac{ik}{4}\sqrt{\frac{4\pi}{3}}\right)\left(\frac{m_2 - m_1}{m_2 + m_1}\right)\bar{R}\langle\psi_{ef}|\left[r_A Y_1^0(\hat{\mathbf{r}}_A) + r_B Y_1^0(\hat{\mathbf{r}}_B)\right]|\psi_{ei}\rangle\mathcal{M}_{r1}^\sigma\mathcal{M}_v^1\Bigg\}\mathcal{M}_{\text{c.m.}}^l$$
$$-\frac{l}{2}\sqrt{\frac{2\pi}{3}}\left(\frac{m_2 - m_1}{m_2 + m_1}\right)\left(\frac{w_{\text{c.m.}}}{w_0}\right)^{|l|-1}\left(\frac{\bar{R}}{w_0}\right)\Big\{\langle\psi_{ef}|\left[r_A Y_1^{\text{sgn}(l)1}(\hat{\mathbf{r}}_A) + r_B Y_1^{\text{sgn}(l)1}(\hat{\mathbf{r}}_B)\right]|\psi_{ei}\rangle\mathcal{M}_{r1}^\sigma\mathcal{M}_v^1$$
$$+\langle\psi_{ef}|\left[r_A Y_1^\sigma(\hat{\mathbf{r}}_A) + r_B Y_1^\sigma(\hat{\mathbf{r}}_B)\right]|\psi_{ei}\rangle\mathcal{M}_{r1}^{\text{sgn}(l)1}\mathcal{M}_v^1\Big\}\mathcal{M}_{\text{c.m.}}^{\text{sgn}(l)(|l|-1)}\Bigg\}, \quad (13)$$

where $\mathcal{M}_{\text{c.m.}}^{l'} = \langle\Psi_{\text{c.m.}f}|(\frac{R_{\text{c.m.}\perp}}{w_{\text{c.m.}}})^{|l'|} e^{il'\Phi_{\text{c.m.}}} e^{ikZ_{\text{c.m.}}}|\Psi_{\text{c.m.}i}\rangle$, $\mathcal{M}_{rl}^m = \langle\psi_{rf}|Y_l^m(\hat{\mathbf{R}})|\psi_{ri}\rangle$, and $\mathcal{M}_v^p = \sum_{s=0}^p \binom{p}{s}(v/\bar{R})^s\langle\psi_{vf}|q^s|\psi_{vi}\rangle$. It is clear from Eqs. (12) and (13) that the c.m. motion can couple with the internal motion in the interaction of the LG beam with a diatomic molecule even within the electric dipole approximation. In the case of homonuclear (nonpolar) molecules, $r = 0$. Thus, Eq. (12) clearly shows that rotational and vibrational motion of nonpolar molecules cannot be influenced by OAM of the LG beam. However, when nonpolar diatomic molecules are polarized and adiabatically or nonadiabatically aligned with a nonresonant intense laser field [35–37], then it is possible for the aligned molecules to interact with an LG beam, leading to the transfer of the light OAM to the rotational motion of the molecules, as we elaborate below.

A particularly interesting prospect for light OAM transfer to molecular electronic motion opens if one makes use of the rotational confinement due to the alignment of internuclear axes of diatomic molecules with a linearly polarized off-resonant intense laser field [35–37]. The alignment and orientation of diatomic molecules and the consequent phenomena of anisotropic ionization [38] and high-order-harmonic generation (HHG) [39–41] have now become an important topic of research from both experimental and theoretical points of view, providing new insight into the underlying effects of rotational confinement on molecular electronic orbitals. Since the rotation of the internuclear axis of a diatomic molecule can couple to the molecular electronic orbital, the interaction of aligned molecules with light carrying OAM will be interesting for OAM transfer in dipole interactions. The alignment of nonpolar ground-state molecules is basically a result of the two-photon off-resonant dipole interaction of molecules. The major advantage of using prealigned molecules would be to get a preferential direction of orientation for angular momentum transfer. If a far-off-resonant and relatively intense LG beam is allowed to interact with prealigned molecules, it is possible to orient the axis of the molecules by the transfer of light orbital angular momentum. Since the molecular axis orientation is coupled to the electronic angular momentum, light OAM will eventually alter the electronic angular momentum. Thus, our theoretically proposed mechanism of light OAM transfer can be experimentally verified by applying an LG beam to diatomic molecules which are already aligned and subsequently probing rotational dynamics with a probe laser via detecting ionization and HHG signals [40]. The light OAM transfer can be inferred by comparing results with and without the LG beam. Thus, with the currently available technology of molecular alignment and orientation with a pump-probe-type setup, it is possible to manipulate molecular rotational and electronic orbital motion with light carrying orbital angular momentum.

In the next section, we discuss numerical results showing the effect of the coupling of c.m. and electronic motions on the quadrupole transition rate in the interaction of an LG beam with ^{23}Na BEC.

III. QUADRUPOLE INTERACTION OF A BEC WITH AN LG BEAM: NUMERICAL RESULTS

In the previous section, we have shown that the quadrupole transition is the lowest-order transition in the interaction of cold atoms with an LG beam for transferring field OAM to electronic motion via quantized c.m. motion. Here we present numerical results showing the effect of the coupling between c.m. and internal motions on quadrupole transition rates in the interaction of an atomic BEC with an LG beam. The first term of Eq. (8) shows this transfer of optical OAM to electronic motion via quantized c.m. motion. This term is important for revealing the predicted effect in quadrupole transitions.

Here we calculate the quadrupole transition rates considering quantum-mechanical and coupled motions of both the electronic and c.m. degrees of freedom of atoms. We consider the c.m. atomic state as the ground state and different vortex states of a trapped BEC [24,42–44]. The system is dilute enough that the Gross-Pitaevskii (GP) theory [45] for trapped bosons is applicable to the c.m. motion. Vortex states in BEC will be created due to the transfer of OAM from the LG beam to the c.m. motion of the atoms. In addition to the OAM, the linear momentum (LM) of light will also be transferred to the c.m. motion of atoms. Hence, we write the initial and final stationary states of the c.m. motion of atoms as [43]

$$\Psi_{\text{c.m.}i}(\mathbf{R}_{\text{c.m.}}) = \psi_{\text{c.m.}i}(R_{\text{c.m.}\perp}, Z_{\text{c.m.}})e^{i\kappa_i\Phi_{\text{c.m.}}} \tag{14}$$

and

$$\Psi_{\text{c.m.}f}(\mathbf{R}_{\text{c.m.}}) = \psi_{\text{c.m.}f}(R_{\text{c.m.}\perp}, Z_{\text{c.m.}})e^{i\kappa_f\Phi_{\text{c.m.}}}e^{ikZ_{\text{c.m.}}}, \tag{15}$$

where κ is the quantum of circulation of atoms about the z axis. $\kappa \neq 0$ represents vortex states of the BEC. The expression of $\mathcal{M}^{l'}_{\text{c.m.}}$ gives the selection rule for the c.m. motion as $\Delta\kappa = l'$. According to time-independent GP theory, the nonlinear Schrödinger equation obeyed by the c.m. motion in an anisotropic simple harmonic potential trap is given by [43,45]

$$\begin{aligned}\Bigg[&-\frac{\hbar^2}{2m_t}\left(\frac{\partial^2}{\partial R^2_{\text{c.m.}\perp}} + \kappa^2 R^{-2}_{\text{c.m.}\perp} + \frac{\partial^2}{\partial Z^2_{\text{c.m.}}}\right)\\ &+ \frac{m_t}{2}\left(\omega^2_\perp R^2_{\text{c.m.}\perp} + \omega^2_Z Z^2_{\text{c.m.}}\right)\\ &+ \frac{4\pi\hbar^2 a}{m_t}|\psi_{\text{c.m.}}|^2\Bigg]\psi_{\text{c.m.}} = \mu\psi_{\text{c.m.}},\end{aligned} \tag{16}$$

where $\omega_\perp$ and ω_Z are the two angular frequencies associated with the external potential of the anisotropic trap, a is the s-wave scattering length, and μ is the chemical potential. In Eq. (8), $w_{\text{c.m.}}$ is of the order of the characteristic length of the trap $a_\perp = (\frac{\hbar}{m_t\omega_\perp})^{1/2}$, and w_e is of the order of the Bohr radius a_0. We evaluate the c.m. wave function at zero temperature using the steepest descent method for functional minimization as prescribed in Ref. [43]. The electronic portion of the transition matrix element is calculated using coupled-cluster theory [34].

We now proceed to numerically evaluate the quadrupole transition rates considering the first term of Eq. (8) where the c.m. and electronic motions are coupled. Let us consider a left-circularly polarized LG beam [$\sigma = +1$ in Eqs. (6)–(8)] with $l = +2$ that is interacting with a BEC with 10^3 ^{23}Na atoms in an anisotropic harmonic trap. The axis of the beam and the axis of the trap are the same and along the z axis of the laboratory frame. Here it is important to note that one unit of the field OAM changes the vorticity of the c.m. wave function and the other unit of field OAM is transferred to the electronic motion via quantized c.m. motion.

For numerical illustration, we choose the characteristics of the experimental trap as given in Ref. [24]. The asymmetry parameter of the trap is $\lambda_{tr} = \omega_Z/\omega_\perp = 2$. The axial frequency $\omega_Z/2\pi = 40$ Hz. The corresponding characteristic length is $a_\perp = 4.673\times10^{-6}$ m. The s-wave scattering length $a = 2.75$ nm [46,47]. The waist of the LG beam is $w_0 = 10^{-4}$ m, and the intensity $I = 10^2$ W cm^{-2}. The amplitude of the LG beam in Eq. (8) is related to the intensity by $I = \epsilon_0 c\epsilon_1^2/2$, where ϵ_0 is vacuum permittivity.

The initial electronic state is $|3S_{\frac{1}{2},\frac{1}{2}}\rangle$. The electronic portion $\langle\psi_f|r^2Y_1^\sigma(\hat{\mathbf{r}})\cos\theta|\psi_i\rangle$ of the second term on the right-hand side of Eq. (8) indicates that the magnetic quantum number of the final electronic state ψ_f will be changed by one unit due to the polarization of light. So the final electronic state will be $|3D_{\frac{3}{2},\frac{3}{2}}\rangle$ or $|3D_{\frac{5}{2},\frac{3}{2}}\rangle$. But the electronic portion $\langle\psi_f|\frac{r^2}{w_e}Y_1^\sigma(\hat{\mathbf{r}})\sin\theta e^{\text{sgn}(l)i\phi}|\psi_i\rangle$ of the first term on the right-hand side of Eq. (8) shows that, in addition to the polarization of the beam, one unit of field OAM can be transferred to the electronic motion via quantized c.m. motion. That increases the magnetic quantum number by two units in the final electronic state, i.e., $|3D_{\frac{5}{2},\frac{5}{2}}\rangle$. Thus, the electronic transition $|3S_{\frac{1}{2},\frac{1}{2}}\rangle \longrightarrow |3D_{\frac{5}{2},\frac{5}{2}}\rangle$ can be realized. Next, we show how quadrupole transition rates are enhanced due to quantized c.m. motion and its coupling with the internal motion.

Figure 2 presents the radial part of the c.m. wave function of the cold atom corresponding to the ground state and different vortex states in the simple harmonic potential trap. The value of the electronic portion of the transition matrix element remains

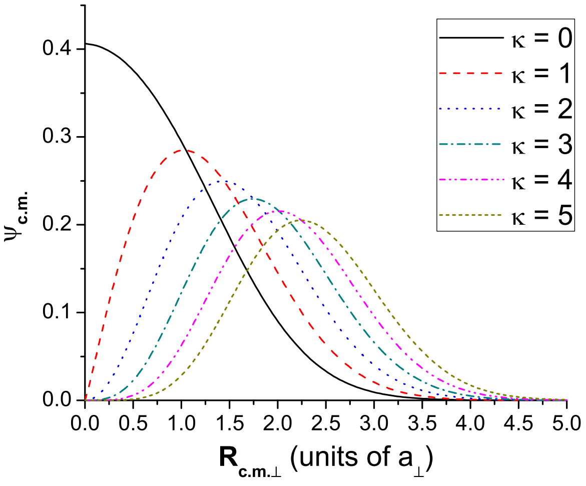

FIG. 2. (Color online) Radial dependence of the c.m. wave function corresponding to the ground state and different vortex states of BEC of ^{23}Na atoms.

unchanged during variation of the vorticity of the c.m. wave function. In the ground state of the c.m. motion, the probability of finding the atom on the beam axis is maximum. But as the vorticity of the states increases, the probability of finding the atom away from the beam axis increases. Hence, the value of the c.m. matrix element $\mathcal{M}^1_{\text{c.m.}}$ increases. This in turn influences the electronic motion due to coupling with the c.m. motion. Table I presents the variation of the c.m. transition matrix element and the quadrupole transition rates W^q with the vorticity of the involved c.m. states. It is clearly shown that with the increase in κ_i the quadrupole transition rate increases due to the increase in the spread of the c.m. wave function. It is worth pointing out that W^q is enhanced because the c.m. motion is treated quantum mechanically and it is coupled with the electronic motion.

Before ending this section, it is worthwhile to mention that, although for simplicity we have neglected the electron's spin, the theoretical treatment we have presented can be extended to include the electron's spin, particularly in situations where the spin-orbit interaction of atoms or molecules is strong. In that case and in the absence of hyperfine interaction, the good quantum number of an atom or molecule will be $\mathbf{J}_e = \mathbf{L}_e + \mathbf{S}$, where $\mathbf{L}_e$ and $\mathbf{S}$ stand for total orbital and spin angular momentum of all the valence electrons of an atom or a molecule. In the case of diatomic molecules, the projection Ω of $\mathbf{J}_e$ on the internuclear axis of the molecule will be a good quantum number. As in atoms and molecules, spin-orbit interaction also arises in light. Circular polarization of light is associated with the spin of a photon. At a fundamental level, it is the vectorial sum of polarization and orbital angular momenta of a photon, which is a conserved quantity. Light OAM will be nearly a good quantum number under paraxial approximation only. In recent times several experiments [48–52] have demonstrated the transfer of angular momentum in light-matter interactions through spin-orbit interactions or interconversion between spin and angular momenta of photons in different physical systems, for example, in nematic liquid crystal [49–51] and nanoplasmonics [48,52]. The transfer of angular momentum in the interaction of light with nematic liquid crystal is quite interesting. The mechanism of light orbital angular momentum transfer presented in this paper is consistent with the method of OAM transfer in liquid crystal as described in Ref. [49].

TABLE I. Variation of $\mathcal{M}^1_{\text{c.m.}}$ (in units of $a_\perp$) and quadrupole transition rates (in 10^3 s^{-1}) with the vorticity of the initial state of the c.m. wave function.

κ_i	κ_f	$\mathcal{M}^1_{\text{c.m.}}$	W^q
0	1	1.000 93	2.48
1	2	1.414 22	4.94
2	3	1.732 05	7.42
3	4	2.000 00	9.89
4	5	2.236 06	12.36
5	6	2.449 48	14.83

IV. CONCLUSION

In conclusion, we have studied the interaction of an LG beam with an atom and a diatomic molecule. We have found that if the atom or molecule is cold enough for its c.m. motion to be quantized, then an angular momentum exchange can take place between the c.m. and internal motions of the system. Atomic and molecular dimensions are far too small compared to the core size of the LG beam. However, if the atom or molecule is cooled down to its recoil limit such that the spread of its c.m. wave function is comparable to the wavelength of the beam, then it can feel the spatial variation of the electric field along the radial direction. The interaction with the LG beam of Eq. (1) ensures that the orientation of the angular momentum transferred from the c.m. motion to the internal motion is the same as that of the field OAM. Our calculations clearly show that the extra angular momentum (other than that coming from the polarization of the field) to the internal motion comes from the quantized c.m. motion. It may be possible to observe this effect with ultracold atoms or molecules interacting with an LG beam as suggested by van Enk [2] 20 years ago. We have numerically calculated the quadrupole transition rates in the interaction of an LG beam with atomic BEC where both the c.m. and electronic motions are quantized and coupled. We have shown the dependence of the transition rates on the vorticity of the c.m. wave function involved in the transition. In the case of molecules at ultracold temperature or in quantum degenerate molecular gases, it would be possible to transfer the field OAM to internal motion even in the dipole interaction. One potential application of the discussed effect can be thought of in quantum information processing using entangled angular momentum observables belonging to the same atom or molecule. Muthukrishnan and Stroud [18] have shown the entanglement between electronic and c.m. degrees of freedom of a cold atom interacting with an LG beam. They have considered the electric dipole interaction where the field OAM couples to the c.m. only and the field polarization couples to electronic motion. If the c.m. motion is allowed to couple with the internal degrees of freedom through a laser-generated synthetic gauge field [53], then it is possible to realize the transfer of light orbital angular momentum into the internal motion in the otherwise similar experimental scenario of Ref. [18]. This will provide a new avenue for entanglement manipulation in angular degrees of freedom. In this context, it is encouraging to note that the entanglement between electronic and vibrational degrees of freedom is already produced for an ion in a linear trap [54], which could be a suitable system to explore the effect predicted in this paper. Our calculations further suggest that the quantized c.m. motion of an atom can couple to electronic motion in the electric quadrupole interaction. The situation is even more interesting in the case of molecules. Equation (12) suggests that the c.m., electronic, rotational, and vibrational motions can be coupled in the interaction with an LG beam even in the electric dipole approximation. We have discussed how the induced polarization and alignment of diatomic molecules due to a linearly polarized intense laser field can be useful for experimental verification of our theory. The alignment and orientation of the molecules of nematic liquid crystal have been found to play an important role in OAM transfer [49–52], indicating that similar effects in gas-phase molecules in intense laser fields will be important for light OAM transfer to the molecules. Finally, the recent advent of ultralong-range cold Rydberg molecules whose electronic orbital can be as large as 100 nm [55,56] and the demonstration of the alignment of such molecules [57] open a new avenue for studying OAM transfer in light-molecule interactions with a huge enhancement of the size effects for the ability of the molecular electrons to experience the spatial variation of light intensity.

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