Mechanism of Angular Momentum Exchange between Molecules and Laguerre-Gaussian Beams

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We derive the interaction Hamiltonian between a diatomic molecule and a Laguerre-Gaussian beam under the assumption of a small spread of the center of mass wave function of the molecule in comparison with the beam waist. Considering the dynamical variables of the center of mass, vibrational, rotational, and electronic motion, we show that, within the electronic dipole approximation, the orbital angular momentum of the field couples with the rotational and electronic motion. The changes in the transition probabilities and selection rules induced by the field orbital angular momentum and the applicability of the derived interaction mechanisms for polyatomic molecules are discussed.

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The interaction of a electromagnetic field with matter has been of great interest since the early years of the development of quantum mechanics. Processes such as emission, absorption, and scattering of electromagnetic field are naturally described in terms of spherical waves [1]. The study of the interaction of matter with fields possessing a spatial profile started with the introduction of lasers beams whose profiles can be shaped, e.g., as Laguerre-Gaussian (LG) or Bessel profile. Recently, the investigation of the properties and the potential applications of beams carrying orbital angular momentum (OAM) has been triggered by the work of Allen and co-workers [2], who have shown that LG beams bear a well-defined amount of OAM per unit energy. The interaction of atoms in free space with strongly focused beams has been studied [3] and, moreover, the challenging task of field bearing OAM interacting with the inner structure of the atoms has been undertaken [4–6], various schemes of OAM exchange between field and atom were identified [4–6], and the entanglement between the external and internal OAM of the atom was shown to emerge [7]. Reference [8] gathers several published papers on both theoretical and experimental results in which the concept of OAM plays a crucial role.

In the present work we study the interaction between a diatomic molecule and a field possessing OAM. The influence of the field OAM on the transition probability and the new established selection rules are discussed. In the end, we make remarks on the applicability of the derived interaction mechanisms to the polyatomic molecules.

We consider a diatomic molecule, e.g., H_2^+ for simplicity, comprising of three particles: two nuclei of mass (charge) m_1 $(+e_1)$ and m_2 $(+e_2)$, and one electron m_e $(-e)$. For simplicity, in the subsequent analysis the spin of the particles is ignored. We shift from an arbitrary coordinate system to the center of mass (c.m.) one and introduce the position vectors $\mathbf{r}_i^{\text{c.m.}} = \mathbf{r}_i - \mathbf{R}$ with $\mathbf{R} = (\sum m_i \mathbf{r}_i)/m_t$ and m_t being the total mass of the molecule. For simplicity we shall drop from now on the index c.m. of the coordinate $\mathbf{r}_i^{\text{c.m.}}$. The nuclei oscillations q are written explicitly in the nuclear and electronic coordinates: $\mathbf{r}_1 = \bar{\mathbf{r}}_1 + \mathbf{v}_1 q$, $\mathbf{r}_e = \mathbf{r} + \bar{\mathbf{r}}_1 + \mathbf{v}_1 q$, where $\bar{\mathbf{r}}_1$ is the equilibrium position of the first nucleus, $\mathbf{v}_1$ is a constant vector of the same orientation as $\mathbf{r}_1$ and of magnitude given by the properties of the molecule, and $\mathbf{r}$ is the relative coordinate of the electron to the nucleus, see Fig. 1.

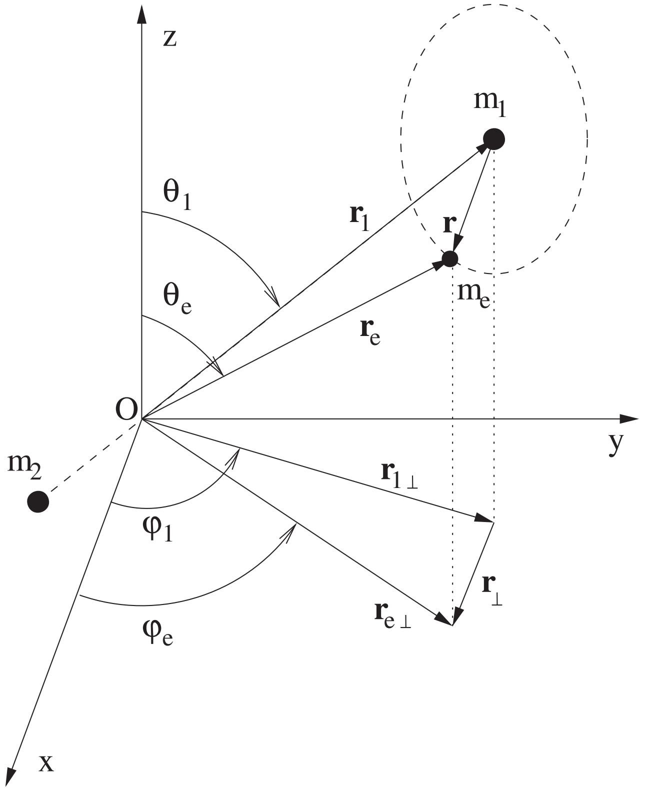

FIG. 1. Diatomic molecule in the center of mass coordinate system: position vectors and their projection onto xOy plane are shown for nucleus m_1 and the electron m_e.

The Hamiltonian of the molecule-field system is given by $H = H_0 + H_{\text{int}}$ where H_0 stands for the unperturbed Hamiltonian of the molecule and H_{int} for their interaction Hamiltonian, specified later in the text. The quantum state of our diatomic molecule is described by the product of four wave functions $|\Psi\rangle = |a\rangle|\nu\rangle|\chi\rangle|\text{c.m.}\rangle$, where $|a\rangle$ stands for electronic state, $|\nu\rangle$ for the vibrational, $|\chi\rangle$ for the rotational, and $|\text{c.m.}\rangle$ for the c.m. wave function, with the associated dynamical variables $\mathbf{r}$, q, $\hat{\mathbf{r}}_1$, and $\mathbf{R}$, respectively. The interaction Hamiltonian is derived in a straightforward manner using the Power-Zienau-Wooley (PZW) scheme [9]:

$$H_{\text{int}} = -\int d^3r\mathcal{P}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}, t), \tag{1}$$

where $\mathcal{P}(\mathbf{r})$ is the electric polarization written in closed integral form [9]:

$$\mathcal{P}(\mathbf{r}) = \sum_n e_n \mathbf{r}_n \int_0^1 \mathrm{d}\lambda \delta(\mathbf{r} - \mathbf{R} - \lambda \mathbf{r}_n). \tag{2}$$

As we are interested in the interaction of the electromagnetic field with molecule via electronic transitions, only the contribution of the electron charge is kept in the expression of the polarization $\mathcal{P}(\mathbf{r})$. Next we assume an electromagnetic field of the LG shape without off-axis radial nodes. In the limit of the spatial small spread of c.m. wave function the molecule experiences a local field of the type $\mathbf{E}(\mathbf{r}_\perp) = \mathbf{E}_0 r_\perp^{|l|} \exp(\mathrm{i}l\varphi)$. A field of this form can be written in terms of regular solid spherical harmonic functions as [10]

$$\mathbf{E}(\mathbf{r}, t) = \mathcal{C}_l \mathbf{E}_0 \mathcal{R}_{|l|}^{l}(\mathbf{r}_\perp) \mathrm{e}^{\mathrm{i}(kz - \omega t)}, \tag{3}$$

with the field normalization constant $\mathcal{C}_l = (-)^{(l+|l|)/2} 2^{|l|} \sqrt{|l|!}$ and $\mathcal{R}_l^m(\mathbf{r}) = \mathcal{N}_{l,m}(r/w_0)^l Y_l^m(\theta, \phi)$ represents the regular solid spherical harmonic function [11]. The normalization constant of the regular solid spherical harmonic function is given by $\mathcal{N}_{l,m} = \sqrt{4\pi/(2l+1)(l+m)!(l-m)!}$. The beam waist w_0 has been explicitly included in the expression of $\mathcal{R}_l^m(\mathbf{r})$ for simplicity. Replacing the expression of the electromagnetic field into Eq. (1) and performing the integral over $\mathrm{d}^3 r$ the interaction Hamiltonian becomes:

$$H_{I,e} = -\mathcal{C}_l e \mathbf{r}_e \cdot \mathbf{E}_0 \int_0^1 \mathrm{d}\lambda \mathcal{R}_{|l|}^{l}(\mathbf{R}_\perp + \lambda \mathbf{r}_{e\perp}) \times \exp(\mathrm{i}[k(R_z + \lambda z_e) - \omega t]) + \text{H.c.} \tag{4}$$

The c.m. and electronic coordinate occurring as arguments of the regular solid spherical harmonic in the above relation are separated using the translation property of the solid spherical harmonics [11]

$$\mathcal{R}_l^m(\mathbf{r}_1 \pm \mathbf{r}_2) = \sum_{l_1=0}^{l} \sum_{m_1=-l_1}^{l_1} (\pm)^{l_1} \mathcal{R}_{l_1}^{m_1}(\mathbf{r}_1) \mathcal{R}_{l-l_1}^{m-m_1}(\mathbf{r}_2). \tag{5}$$

In the previous works similar procedures were employed, e.g., Babiker and co-workers [5] used a power expansion formula to split the c.m. and "electronic-type" coordinates assuming that the modulus of c.m. coordinate is much smaller than the electronic-type one, while in the analysis of Bessel beams interacting with atoms [6] coordinate splitting formulas were provided by addition theorems of Bessel functions [12]. The integral over λ is performed while writing $\exp(\mathrm{i}\lambda k z_e) \simeq 1 + \mathrm{i}\lambda k z_e$ and the interaction Hamiltonian reads:

$$H_{I,e} = -\mathcal{C}_l e \mathbf{r}_e \cdot \mathbf{E}_0 \mathrm{e}^{\mathrm{i}kR_z} \sum_{l_1=0}^{|l|} \sum_{m_1=-l_1}^{l_1} \left(\frac{1}{|l| - l_1 + 1} + \frac{\mathrm{i}k z_e}{|l| - l_1 + 2} \right) \mathcal{R}_{l_1}^{m_1}(\mathbf{R}_\perp) \mathcal{R}_{|l|-l_1}^{l-m_1}(\mathbf{r}_{e\perp}) + \text{H.c.}, \tag{6}$$

where the time dependence has been dropped for simplicity. In Eq. (6) the terms contributing to the electronic interaction are the overall factor $\mathbf{r}_e$, the term within the round brackets and $\mathcal{R}_{|l|-l_1}^{l-m_1}(\mathbf{r}_{e\perp})$. Next we replace the expression of the electronic coordinates (in the c.m. reference frame) by $\mathbf{r}_e = \mathbf{r} + \mathbf{r}_1$ and split the solid regular spherical harmonic function which has the electronic coordinate as the argument, according to Eq. (5). From the emerging terms we keep only those which give rise to electronic dipole interaction and $H_{I,e}$ becomes

$$\begin{aligned} H_{I,e}^{(d)} = -\mathcal{C}_l \mathrm{e}^{\mathrm{i}kR_z} \Bigg\{ & e\mathbf{r} \cdot \mathbf{E}_0 \sum_{l_1=0}^{|l|} \sum_{m_1=-l_1}^{l_1} \left[\frac{1}{|l| - l_1 + 1} + \frac{\mathrm{i}k r_1 \cos\theta_1}{|l| - l_1 + 2} \right] \mathcal{R}_{l_1}^{m_1}(\mathbf{R}_\perp) \mathcal{R}_{|l|-l_1}^{l-m_1}(\mathbf{r}_{1\perp}) \\ & + e\mathbf{r}_1 \cdot \mathbf{E}_0 \sum_{l_1=0}^{|l|-1} \sum_{m_1=-l_1}^{l_1} \left[\frac{1}{|l| - l_1 + 1} + \frac{\mathrm{i}k r_1 \cos\theta_1}{|l| - l_1 + 2} \right] \mathcal{R}_{l_1}^{m_1}(\mathbf{R}_\perp) \sum_{m_2=-1}^{+1} \mathcal{R}_1^{m_2}(\mathbf{r}_\perp) \mathcal{R}_{|l|-l_1-1}^{l-m_1-m_2}(\mathbf{r}_{1\perp}) \\ & + erY_1^0(\theta, \varphi) \mathbf{r}_1 \cdot \mathbf{E}_0 \sum_{l_1=0}^{|l|} \sum_{m_1=-l_1}^{l_1} \frac{\mathrm{i}k}{|l| - l_1 + 2} \mathcal{R}_{l_1}^{m_1}(\mathbf{R}_\perp) \mathcal{R}_{|l|-l_1}^{l-m_1}(\mathbf{r}_{1\perp}) \Bigg\} + \text{H.c.} \end{aligned} \tag{7}$$

In the second double sum of the above relation, the term of index $l_1 = |l|$ has been discarded because it is proportional to $\sum_{m_1=-|l|}^{|l|} \mathcal{R}_{|l|}^{m_1}(\mathbf{R}_\perp) \mathcal{R}_0^{l-m_1}(\mathbf{r}_{1\perp}) = \mathcal{R}_{|l|}^{l}(\mathbf{R}_\perp) \mathcal{R}_0^0(\mathbf{r}_{1\perp})$ and, therefore, it cannot mediate electronic dipole transitions, but only c.m. ones. Since all solid regular spherical harmonic functions occurring in relation (7) have as argument vectors which are perpendicular to axis Oz, i.e., $\mathcal{R}_l^m(\mathbf{r}_\perp) \sim r^l Y_l^m(\theta = \pi/2, \phi)$, we employ the properties of spherical harmonics as well as those of double factorial to cast relation (7) into a simpler form. For instance, considering the first double sum occurring in the right-hand side of relation: the double sum over l_1 and m_1 reduces to a single sum over l_1 with $m_1 = \mathrm{sgn}\,(l)l_1$, as it was shown in Ref. [10]. The products of the type $\mathbf{r}_i \cdot \mathbf{E}_0$ will be replaced by $r_i\sqrt{4\pi/3}\sum_{\sigma=\pm1}\epsilon_\sigma Y_1^\sigma(\theta_i, \varphi_i)$, σ being associated with the polarization of the electric field, $\epsilon_{\pm1} = (E_x \pm \mathrm{i}E_y)/\sqrt{2}$. The interaction Hamiltonian will read as:

$$H_{I,e}^{(d)} = -\frac{16\pi^2}{3}\mathcal{C}_l e r \mathrm{e}^{\mathrm{i}kR\cos\Theta}\sum_{\sigma=\pm1}\sum_{l_1=0}^{|l|}\frac{\epsilon_\sigma}{[(2l_1+1)!]^{1/2}}\left(\frac{R}{w_0}\right)^{l_1}Y_{l_1}^{\mathrm{sgn}\,(l)l_1}(\hat{\mathbf{R}})\left\{S_1\left(\frac{r_1}{w_0}\right)^{|l|-l_1}Y_{|l|-l_1}^{\mathrm{sgn}\,(l)(|l|-l_1)}(\hat{\mathbf{r}}_1)Y_1^\sigma(\hat{\mathbf{r}})\right.$$
$$\left.+ S_2\left(\frac{r_1}{w_0}\right)^{|l|-l_1}Y_1^\sigma(\hat{\mathbf{r}}_1)Y_{|l|-l_1-1}^{\mathrm{sgn}\,(l)(|l|-l_1-1)}(\hat{\mathbf{r}}_1)Y_1^{\mathrm{sgn}\,(l)}(\hat{\mathbf{r}}) + S_3\left(\frac{r_1}{w_0}\right)^{|l|-l_1+1}Y_1^\sigma(\hat{\mathbf{r}}_1)Y_{|l|-l_1}^{\mathrm{sgn}\,(l)(|l|-l_1)}(\hat{\mathbf{r}}_1)Y_1^0(\hat{\mathbf{r}})\right\} + \mathrm{H.c.}, \tag{8}$$

where

$$S_{\{{1 \atop 2}\}} = \left(\frac{1}{|l|-l_1+1} + \frac{\mathrm{i}kr_1\cos\theta_1}{|l|-l_1+2}\right) \times \left[\begin{Bmatrix} 3/4\pi \\ 1/2 \end{Bmatrix}\frac{1}{\Gamma[2(|l|-l_1)+\{{2 \atop 0}\}]}\right]^{1/2} \tag{9a}$$

$$S_3 = \frac{\mathrm{i}kw_0}{|l|-l_1+2}\sqrt{\Gamma[2(|l|-l_1+1)]}, \tag{9b}$$

with $S_2 = 0$ for $l_1 = |l|$. The interaction Hamiltonian (8) represents the main result of our work. It allows us to identify three mechanisms of angular momentum exchange between c.m., rotational, and electronic motion, all of them interceded by electronic dipole transition: (i) the field polarization σ couples to the electronic motion and the field orbital angular momentum is imparted between c.m. and rotational motion of molecule, (ii) the field polarization couples to the rotational motion of molecule and the field OAM is imparted between c.m., rotational, and electronic motion, and (iii) the field polarization couples to the rotational motion, the field OAM is imparted between c.m. and rotational motion with the electronic motion "assisting" the OAM exchange, while couples neither to field polarization, nor to field OAM. The second mechanism indicates that, contrary to previous results [5,10], the field OAM couples in the paraxial approximation to the electronic motion even in the electronic dipole interaction. In all above mechanisms the field polarization couples only to the internal degrees of freedom of the molecule, while the field OAM couples to both internal and external degrees of freedom, a fact consistent with the frame of paraxial approximation. Each term in the curl brackets of relation (8) comes with a factor r_1/w_0. Since $r_1 \ll w_0$, or equivalently $kr_1 \ll 1$, the third interaction mechanism and the terms kr_1 occurring in the coefficients $S_{1,2}$ will have smaller contribution to the total transition probability. In the subsequent analysis we will discard these higher order terms. Now we proceed with the evaluation of the transition matrix element $\mathcal{M}_{i\to f} = \langle\Psi_f|H_{I,e}^{(d)}|\Psi_i\rangle$:

$$\mathcal{M}_{i\to f} = -\frac{16\pi^2}{3}\mathcal{C}_l e\sum_{\sigma=\pm1}\sum_{l_1=0}^{|l|}\frac{\epsilon_\sigma}{[(2l_1+1)!]^{1/2}}\left(\frac{w_R}{w_0}\right)^{l_1}\left(\frac{\bar{r}_1}{w_0}\right)^{|l|-l_1}\mathcal{M}_{\mathrm{c.m.}}^{(l_1)}\mathcal{M}_\nu^{(|l|-l_1)}[S_1\mathcal{M}_\chi^{(0,|l|-l_1)}\langle f|rY_1^\sigma(\hat{\mathbf{r}})|i\rangle$$
$$+ S_2\mathcal{M}_\chi^{(\sigma,|l|-l_1-1)}\langle f|rY_1^{\mathrm{sgn}\,(l)}(\hat{\mathbf{r}})|i\rangle], \tag{10}$$

where w_R stands for the spatial spread of the c.m. wave function and

$$\mathcal{M}_{\mathrm{c.m.}}^{(l_1)} = \langle\mathrm{c.m.}_f|\left(\frac{R}{w_R}\right)^{l_1}Y_{l_1}^{\mathrm{sgn}\,(l)l_1}(\hat{\mathbf{R}})\mathrm{e}^{\mathrm{i}kR_z\cos\Theta}|\mathrm{c.m.}_i\rangle \tag{11a}$$

$$\mathcal{M}_\nu^{(p)} = \sum_{s=0}^{p}\binom{p}{s}(v_{1i}/\bar{r}_1)^s\langle\nu_f(\bar{r}_f)|q^s|\nu_i(\bar{r}_i)\rangle \tag{11b}$$

$$\mathcal{M}_\chi^{(a,b)} = \langle\chi_f|Y_1^a(\hat{\mathbf{r}}_1)Y_b^{\mathrm{sgn}\,(l)b}(\hat{\mathbf{r}}_1)|\chi_i\rangle. \tag{11c}$$

The matrix elements (11) establish the selection rules and the probability of the electronic transitions governed by the interaction Hamiltonian (8). One remarks that the contribution of each term to the total sum occurring in Eq. (10) is multiplied by $(w_R/w_0)^{l_1}$ and $(\bar{r}/w_0)^{|l|-l_1}$ and, therefore, the order of magnitude of these ratios is important when deriving transition probabilities. The changes in electronic configuration, due to the electronic transition, will induce modification into the oscillatory motion of the nuclei. Therefore, in the expression of the vibrational transition matrix element (11b) the dependence on the internuclear separation $\bar{\mathbf{r}}$ of the final and initial vibrational states has been written explicitly in the case of the harmonic potential. Since the electronic transitions take place practically at constant internuclear distance, according to Frank-Condon principle, the electronic dipole interaction Hamiltonian (8) includes $\bar{\mathbf{r}}_i$, i.e., $\bar{\mathbf{r}}_{1i} \propto \bar{\mathbf{r}}_i$, as parameter and not as dynamical variable. Considering the case of no OAM, i.e., $l = 0$ implying $p = 0$ in relation (11b), the vibrational matrix element $\langle\nu_f(\bar{r}_f)|\nu_i(\bar{r}_i)\rangle$, possessing analytic expression [13], can link vibrational states of the same parity only, leading to the change in the vibrational number of the type $\Delta\nu = 0, \pm2, \pm4\ldots$. However, when the field driving the electronic transition bears OAM the vibrational, matrix element can link states of different parities: the odd and even powers of q in formula (11b) will lead to $\Delta\nu = \pm 1, \pm 3, \pm 5\ldots$ and $\Delta\nu = 0, \pm 2, \pm 4\ldots$, respectively. The relation (11c) describes the angular momentum (orbital and polarization) exchange between rotational motion and the field. In comparison with the standard rotational selection rules of the electronic spectroscopy where the change of angular momentum is given by $\Delta\chi = 0, \pm 1$, the use of fields bearing OAM leads to a wider range of $\Delta\chi$, which can be easily computed casting the rotational matrix element in terms of Clebsch-Gordon coefficients. Therefore, when electronic spectroscopy is performed using fields carrying OAM one will expect a widening of the vibrational and rotational spectra.

In the previous c.m. and electronic quadrupole interaction schemes [5,10] the amount of OAM transferred to the c.m motion is fixed to $|l| - 1$ and the total transition probability is of the form $\mathcal{P}_T^{(q)} \propto \mathcal{P}_e^{(q)}(w_R/w_0)^{2(|l|-1)} \times \mathcal{P}_{\text{c.m.}}^{(|l|-1)}$, where $\mathcal{P}$ stands for the c.m. and electronic quadrupole transition probability. Taking into account the maximum values (the highest probability transitions) of the vibrational (11b) and rotational (11c) matrix elements one remarks that they take on values in a restricted range and, therefore, they have almost constant contribution to the total transition probability. Hence, for the interaction scheme described by relation (10), the total transition probability is roughly given by $\mathcal{P}_T^{(d)} \propto \mathcal{P}_e^{(d)} \sum_{l_1=0}^{|l|-1} (w_R/w_0)^{2l_1} (\bar{r}/w_0)^{2(|l|-l_1)} \mathcal{P}_{\text{c.m.}}^{(l_1)}$. All terms are kept because, depending on the given parameters, only a specific value of l_1 will yield significant contribution to the total probability.

For polyatomic molecules the nuclear coordinate $\mathbf{r}_1$ includes the normal oscillation modes q_j in the form $\mathbf{r}_1 = \bar{\mathbf{r}}_1 + \sum_j \mathbf{v}_{1j} q_j$, where the vector set $\mathbf{v}_{1j}$ is constant in time and it is determined solely by the structure of the molecule. In this case, the oscillations of nuclei may be described in terms of a two- or three-dimensional harmonic oscillator denoting $\mathbf{Q} = \sum_j \mathbf{v}_{1j} q_j$. In Eq. (7) the factor which conains the rotational-vibrational dynamical variables, i.e., $\hat{\bar{\mathbf{r}}}_1$ and q_j, can be written according to relation (5) as $\mathcal{R}_{l_0}^{m_0}(\mathbf{r}_{1\perp}) = \sum_{l_2,m_2} \mathcal{R}_{l_2}^{m_2}(\bar{\mathbf{r}}_{1\perp}) \mathcal{R}_{l_0-l_2}^{m_0-m_2}(\mathbf{Q}_{1\perp})$, with l_0 and m_0 taking on the values of the corresponding lower and upper index, and where the double sum over (l_2, m_2) can be reduced to a simple sum [10]. The sums occurring in the final expression [14] of the interaction Hamiltonian will include terms of the form $\mathcal{R}_{l_2}^{\text{sgn}\,(l_0)l_2}(\mathbf{Q}) \sim Q_1^{l_2} Y_{l_2}^{\text{sgn}\,(l_0)l_2}(\hat{\mathbf{Q}})$, which indicates that the field OAM couples to the vibrational motion of nuclei as well. However, in general, the probability that such process will occur is expected to be small due to the factor $(w_1/w_0)^{l_2}$ where w_1 stands for the spatial spread of the oscillatory motion of the nucleus.

In conclusion, we have derived using analytical calculation the interaction Hamiltonian between a diatomic molecule and an LG beam. Contrary to the previous results, we have shown that the electronic motion and the electromagnetic field can exchange one unit of OAM within electronic dipole interaction. From the structure of the interaction Hamiltonian we have identified OAM exchange schemes between electromagnetic field, on one hand, and c.m. motion, rotational, and electronic motion of the molecules, on the other hand. When using fields carrying OAM in electronic spectroscopy new selection rules emerge and a widening of the vibrational and rotational spectra is expected. The extension of the interacting scenarios to the case of polyatomic molecules is discussed.

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