Orbital Angular Momentum Exchange in the Interaction of Twisted Light with Molecules

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In the interaction of molecules with light endowed with orbital angular momentum, an exchange of orbital angular momentum in an electric dipole transition occurs only between the light and the center of mass motion; i.e., internal "electronic-type" motion does not participate in any exchange of orbital angular momentum in a dipole transition. A quadrupole transition is the lowest electric multipolar process in which an exchange of orbital angular momentum can occur between the light, the internal motion, and the center of mass motion. This rules out experiments seeking to observe exchange of orbital angular momentum between light beams and the internal motion in electric dipole transitions.

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During the past decade or so, the orbital angular momentum (OAM) associated with certain types of laser light has been the focus of much attention in both theoretical and experimental contexts. Interest evolved from work by Allen *et al.* [1], who showed that Laguerre-Gaussian light carries OAM in discrete units of $\hbar$ associated with the azimuthal phase dependence of the field distribution. Since then, a number of experiments have demonstrated the influence which the OAM of light imparts on polarizable matter, leading to novel features, such as the optical spanner effect [2–4]. The manifestation of OAM in interactions of Laguerre-Gaussian light with atoms has been explored theoretically, leading to predictions of a light-induced torque which can be used to control the rotational motion of atoms and ions [5]. Berry [6] showed theoretically that the OAM is an intrinsic property of all types of azimuthal phase-bearing light, independent of the choice of axis about which it is defined. A review of the work carried out until 1999 is given in Ref. [7].

More recently, O'Neil *et al.* [8] investigated the classification of OAM in terms of intrinsic and extrinsic types in the context of Laguerre-Gaussian light, and Muthukrishnan and Stroud [9] explored the entanglement of internal and external angular momenta in a single atom. There is also a very recent report on the measurement of optical OAM [10]. It can be argued that, if orbital angular momentum is indeed an intrinsic property of light, then in its interaction with a bound system of charges such as an atom or a molecule, an exchange should arise between light and matter, especially in a transition between the energy levels, just as the photon spin angular momentum manifests itself in the interaction of circularly polarized light in a radiative transition. The purpose of this article is to explore the validity of this argument using a prototypical model of a molecule interacting with a simple form of light carrying OAM.

We focus on an electromagnetic light mode of frequency ω and orbital angular momentum $l\hbar$, with an electric field vector distribution expressible in cylindrical polar coordinates $\mathbf{r} = (\mathbf{r}_\parallel, z) \equiv (r_\parallel, \phi', z)$ as follows [11]:

$$\mathbf{E}_{kl}(\mathbf{r}, t) = \hat{\boldsymbol{\epsilon}} F(r_\parallel) e^{i(kz-\omega t)} e^{il\phi'}, \tag{1}$$

where $\hat{\boldsymbol{\epsilon}}$ is a wave polarization vector and $F(r_\parallel)$ is a scalar distribution function depending only on the radial coordinate. For the molecule we consider a bound system of charges in simplest form, namely, a hydrogenic two-particle system consisting of a spinless electron (referred to as $e-$) of mass m_1 and charge $-e$ and a spinless nucleus (referred to as $e+$) of mass m_2 and charge $+e$, with e the magnitude of electron charge. It is straightforward to derive the Power-Zienau-Woolley (PZW) Hamiltonian of this system in interaction with the light field [12,13], expressible as a sum of four parts:

$$H = H_M^0 + H_\mu^0 + H_{\text{fields}}^0 + H_{\text{int}}. \tag{2}$$

$H_M^0 = P^2/2M$ is the center of mass Hamiltonian, which is essentially its kinetic energy operator, with $\mathbf{P}$ the center of mass momentum and $M = m_1 + m_2$ its total mass. The center of mass momentum is conjugate to the center of mass coordinate $\mathbf{R}$, defined in terms of the particle position vectors $\mathbf{q}_i$; $i = 1, 2$, by $\mathbf{R} = (m_1\mathbf{q}_1 + m_2\mathbf{q}_2)/M$. Figure 1 schematically shows the position vectors $\mathbf{q}_1$ and $\mathbf{q}_2$ and that of the center of mass $\mathbf{R}$. We are, however, interested in the possibility of the center of mass rotating about a beam axis in which case we should write

$$H_M^0 = \frac{L_z^2}{2I} + \frac{P_z^2}{2M}, \tag{3}$$

where L_z is the angular momentum operator, I is the moment of inertia of the atomic center of mass about the z axis, and P_z is the center of mass momentum axial vector component. The second term in Eq. (2) pertains to the internal "electronic-type" motion,

$$H_\mu^0 = \frac{p^2}{2\mu} - \frac{e^2}{4\pi\epsilon_0 q}, \tag{4}$$

where $\mu = m_1 m_2/M$ is the reduced mass and $\mathbf{p}$ is the

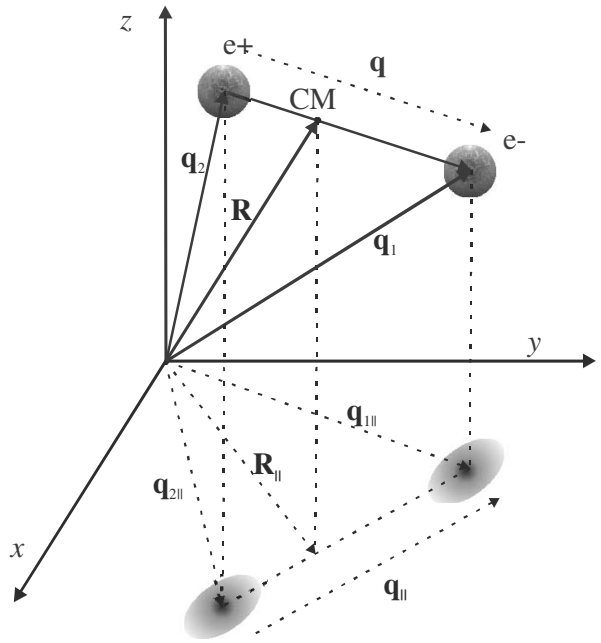

FIG. 1. The particle position vectors and that of the center of mass for the two-particle model of the molecule. The projections of these vectors in the x-y plane are also shown.

momentum conjugate to the internal coordinate $\mathbf{q} = \mathbf{q}_1 - \mathbf{q}_2$. The second term in Eq. (4) is the Coulomb potential binding the two-particle system, with $q = |\mathbf{q}|$. The third term in the total Hamiltonian is defined by

$$H_{\text{field}} = \hbar\omega a_{kl}^{\dagger} a_{kl}, \tag{5}$$

which is the field Hamiltonian in quantized form with a_{kl} the annihilation operator of the light mode of frequency ω, orbital angular momentum $l\hbar$, and axial wave vector $\mathbf{k} = k\hat{\mathbf{z}}$. The validity of such quantization for beams with OAM has recently been vindicated in work by Dávila Romero *et al.* [11]. Finally, the last term in (2) is the Hamiltonian describing the coupling between the light and matter. In the PZW scheme, this can be written as [12,13]

$$H_{\text{int}} = -\int d^3\mathbf{r}\ \mathcal{P}(\mathbf{r}) \cdot \tilde{\mathbf{E}}_{kl}(\mathbf{r}, t), \tag{6}$$

where $\tilde{\mathbf{E}}_{kl}(\mathbf{r}, t)$ is the second quantized form of the electric field in Eq. (1); $\mathcal{P}(\mathbf{r})$ is the electric polarization defined in closed integral form by

$$\mathcal{P}(\mathbf{r}) = \sum_{\alpha=1,2} e_\alpha \int_0^1 d\lambda (\mathbf{q}_\alpha - \mathbf{R}) \delta[\mathbf{r} - \mathbf{R} - \lambda(\mathbf{q}_\alpha - \mathbf{R})]. \tag{7}$$

For simplicity, we have ignored all magnetic interactions. Note that, although the electric polarization field defined in Eq. (7) appears to be a function of $\mathbf{q}_1$ and $\mathbf{q}_2$, it can be written entirely in terms of the relative coordinate $\mathbf{q}$ using

$$\mathbf{q}_{1,2} - \mathbf{R} = \pm m_{2,1}\mathbf{q}/M. \tag{8}$$

We start by specifying zero-order states of the overall motion, comprising the center of mass motion (rotational and translational), the internal "electronic-type" motion, and the field state. The appropriate states are product states of the three-subsystem Hamiltonian $H^0 = H_M^0 + H_\mu^0 + H_{\text{field}}^0$ expressible as $|P_z, L_z; j; \{N_{kl}\}\rangle$. The unperturbed center of mass motion in this state is represented by an axial translational state with linear momentum P_z, together with a rotational eigenstate of the angular momentum operator L_z with eigenvalues $\hbar L_z$. The internal motion enters in terms of the hydrogenic excited discrete states $|j\rangle \equiv |e\rangle$ of energy E_e and a ground state $|j\rangle \equiv |g\rangle$ of energy E_g. The notation $|e\rangle$ and $|g\rangle$ stand for $|n_e; l_e; m_e\rangle$ and $|n_g; l_g; m_g\rangle$, respectively, where n_j, l_j, m_j with $j \equiv e, g$ are hydrogenic state quantum numbers. Finally, the ket$|\{N_{kl}\}\rangle$ is the number state of the light field.

The coupling between matter and field invokes the interaction matrix element $\mathcal{M}_{if}$, where $|i\rangle \equiv |P_z, L_z; e; \{N_{kl}\}\rangle$ and $|f\rangle \equiv |P_z', L_z'; g; \{N_{kl}'\}\rangle$. Specifically,

$$\mathcal{M}_{if} = -\langle P_z, L_z; e; \{N_{kl}\}| \int d^3\mathbf{r} \mathcal{P}(\mathbf{r}) \cdot \tilde{\mathbf{E}}_{kl}(\mathbf{r}, t) |P_z', L_z'; g; \{N_{kl}'\}\rangle. \tag{9}$$

To proceed, we express Eq. (6) as follows:

$$\begin{aligned} H_{\text{int}} &= e \int d^3\mathbf{r} \int_0^1 d\lambda \left\{ \frac{m_2}{M}\mathbf{q}\delta\left[\mathbf{r} - \mathbf{R} - \lambda\frac{m_2}{M}\mathbf{q}\right] + \frac{m_1}{M}\mathbf{q}\delta\left[\mathbf{r} - \mathbf{R} + \lambda\frac{m_1}{M}\mathbf{q}\right] \right\} \cdot \tilde{\mathbf{E}}_{kl}(\mathbf{r}, t) \\ &= \frac{e}{M}\mathbf{q} \cdot \int_0^1 d\lambda \{ m_2 \tilde{\mathbf{E}}_{kl}(\mathbf{R} + \lambda m_2\mathbf{q}/M, t) + m_1 \tilde{\mathbf{E}}_{kl}(\mathbf{R} - \lambda m_1\mathbf{q}/M, t) \} = H_{\text{int}}^{(1)} + H_{\text{int}}^{(2)}, \end{aligned} \tag{10}$$

where we have used Eqs. (7) and (8) and the electric field is that given by Eq. (1), evaluated at $\mathbf{r} = \mathbf{R} + \lambda m_2\mathbf{q}/M$ in $H_{\text{int}}^{(1)}$ and at $\mathbf{r} = \mathbf{R} - \lambda m_1\mathbf{q}/M$ in $H_{\text{int}}^{(2)}$. The most useful form of the interaction Hamiltonian in Eq. (10) is a multipolar. It should, however, be emphasized that, while we can treat the magnitude of the internal coordinate as small relative to a wavelength, we cannot say that the internal azimuthal angle is small, or set it equal to that of the center of mass. We need to incorporate the full azimuthal angular dependence which must be split into internal and center of mass dependences. To establish the azimuthal angular dependence, we consider projections of relevant vectors in the x-y plane. Figure 2 shows the vectors $\mathbf{R}_\parallel$, $\lambda m_2\mathbf{q}_\parallel/M$ and their sum $\mathbf{R}_\parallel + \lambda m_2\mathbf{q}_\parallel/M$ in the context of $H_{\text{int}}^{(1)}$. A similar figure can be constructed for the

vectors $\mathbf{R}_\parallel$; $-\lambda m_1 \mathbf{q}_\parallel/M$ and their sum $\mathbf{R}_\parallel - \lambda m_1 \mathbf{q}_\parallel/M$ in the context of $H^{(2)}_{\rm int}$.

Concentrating on $H^{(1)}_{\rm int}$ first, the azimuthal angles associated with the relevant vector projections as shown in Fig. 2 are as follows: Φ is the azimuthal angle of the vector $\mathbf{R}_\parallel + \lambda m_2 \mathbf{q}_\parallel/M$, while Φ_R is that for the center of mass coordinate $\mathbf{R}_\parallel$ and ϕ is the internal azimuthal angle, which is also the azimuthal angle of the vector $\lambda m_2 \mathbf{q}_\parallel/M$. The sine rule immediately gives

$$\frac{M \sin(\Phi - \Phi_R)}{m_2 \lambda q_\parallel} = \frac{\sin(\phi - \Phi)}{R_\parallel}, \tag{11}$$

which, after expanding the sines, yields

$$\tan(\Phi) = \frac{\lambda m_2 q_\parallel \sin(\phi) + M R_\parallel \sin(\Phi_R)}{\lambda m_2 q_\parallel \cos(\phi) + M R_\parallel \cos(\Phi_R)}. \tag{12}$$

Substituting for $\tilde{\mathbf{E}}_{kl}$ and making use of Eq. (1), we have

$$H^{(1)}_{\rm int} = \frac{e m_2}{M} \hat{\boldsymbol{\epsilon}} \cdot \mathbf{q} \int_0^1 d\lambda F(|\mathbf{R}_\parallel + \lambda m_2 \mathbf{q}_\parallel/M|) e^{ik(R_z + \lambda m_2 q_z/M)} e^{il\Phi} e^{-i\omega t} a_{kl} + \text{H.c.}, \tag{13}$$

where H.c. is the Hermitian conjugate. The azimuthal exponential factor in Eq. (13) is

$$e^{il\Phi} = \left(\frac{[\lambda m_2 q_\parallel \cos(\phi) + M R_\parallel \cos(\Phi_R)] + i[\lambda m_2 q_\parallel \sin(\phi) + M R_\parallel \sin(\Phi_R)]}{\sqrt{[\lambda m_2 q_\parallel \cos(\phi) + M R_\parallel \cos(\Phi_R)]^2 + [\lambda m_2 q_\parallel \sin(\phi) + M R_\parallel \sin(\Phi_R)]^2}} \right)^l. \tag{14}$$

This can be simplified to give

$$e^{il\Phi} = \left(\frac{\lambda m_2 q_\parallel e^{i\phi} + M R_\parallel e^{i\Phi_R}}{\sqrt{[\lambda m_2 q_\parallel]^2 + M^2 R_\parallel^2 + 2 m_2 M \lambda q_\parallel R_\parallel \cos(\phi - \Phi_R)}} \right)^l. \tag{15}$$

We now make use of the "multipolar" approximation,

$$\lambda m_2 q_\parallel/M \ll R_\parallel; \qquad \lambda m_2 q_z/M \ll R_z. \tag{16}$$

These facilitate the next steps starting from Eq. (13) involving the expansion around $R_\parallel$ of the function $F(|\mathbf{R}_\parallel + \lambda m_2 \mathbf{q}_\parallel/M|)$, together with the expansion of the exponential term containing z components around R_z. A similar treatment is needed to expand the azimuthal factor in Eq. (15). These steps are followed by integration over λ. We obtain from Eq. (13) using $\hat{\mathbf{q}}\cdot_\parallel \hat{\mathbf{R}}_\parallel = \cos(\phi - \phi_R)$, where carets denote unit vectors,

$$H^{(1)}_{\rm int} \approx \frac{e m_2}{M} \hat{\boldsymbol{\epsilon}} \cdot \mathbf{q} e^{ikR_z} \left(1 + i \frac{m_2 k q_z}{2M}\right) e^{-i\omega t} a_{kl} \times \left\{ F(R_\parallel) e^{il\phi_R} + \frac{q_\parallel m_2}{M} \left[G_l^-(R_\parallel) e^{i(l-1)\Phi_R} e^{i\phi} + G_l^+(R_\parallel) e^{i(l+1)\Phi_R} e^{-i\phi} \right] \right\} + \text{H.c.}, \tag{17}$$

the functions $G_l^\pm(R_\parallel)$ arising from differentiating $F(R_\parallel)$ with respect to $R_\parallel$, and expanding $e^{il\Phi}$,

$$G_l^\pm(R_\parallel) = \frac{1}{4} \left(\frac{dF}{dR_\parallel} \mp \frac{lF}{R_\parallel} \right). \tag{18}$$

Following similar steps, we can reduce the expression for $H^{(2)}_{\rm int}$ to the analogue of Eq. (17):

$$H^{(2)}_{\rm int} \approx \frac{e m_1}{M} \hat{\boldsymbol{\epsilon}} \cdot \mathbf{q} e^{ikR_z} \left(1 - i \frac{m_1 k q_z}{2M}\right) e^{-i\omega t} a_{kl} \times \left\{ F(R_\parallel) e^{il\phi_R} - \frac{q_\parallel m_1}{M} [G_l^-(R_\parallel) e^{i(l-1)\Phi_R} e^{i\phi} + G_l^+(R_\parallel) e^{i(l+1)\Phi_R} e^{-i\phi}] \right\} + \text{H.c.} \tag{19}$$

The total interaction Hamiltonian is the sum of the expressions in Eqs. (17) and (19). The interaction Hamiltonian in the electric dipole approximation emerges from the sum of the terms linear in the vector components of the internal coordinate $\mathbf{q}$. We have

$$H_{\rm int}(\text{dipole}) = e \hat{\boldsymbol{\epsilon}} \cdot \mathbf{q} e^{ikR_z} F(R_\parallel) e^{il\phi_R} e^{-i\omega t} a_{kl} + \text{H.c.}, \tag{20}$$

and we see that, besides the internal operator $e\hat{\boldsymbol{\epsilon}} \cdot \mathbf{q}$, the dipole approximation involves only the center of mass cylindrical coordinates $(R_\parallel, \Phi_R, R_z)$. Substitution of this in Eq. (9), writing the explicit forms of the translational and rotational eigenstates of the center of mass motion, and performing the space integrals, we obtain

$$\mathcal{M}_{if} = (2\pi)^2 \langle e|\hat{\boldsymbol{\epsilon}} \cdot \mathbf{d}|g\rangle N_{kl}^{1/2} e^{-i\omega t} \delta_{(L_z - L_z'),l} \times \delta(P_z - P_z' - \hbar k) \mathcal{M}_{,\parallel} \tag{21}$$

where $\mathbf{d} = e\mathbf{q}$ is the electric dipole moment vector and $\mathcal{M}_\parallel$ is the integral $\mathcal{M}_\parallel = \int_0^\infty dR_\parallel R_\parallel F(R_\parallel)$. The Dirac delta function in Eq. (21) exhibits conservation of the center of mass axial linear momentum with conventional linear momentum transfer between the light and the center of mass. The Kronecker delta expresses conservation of orbital angular momentum; there is clearly OAM transfer of magnitude $l\hbar$ between the light and the center of mass rotational motion. In this electric dipole

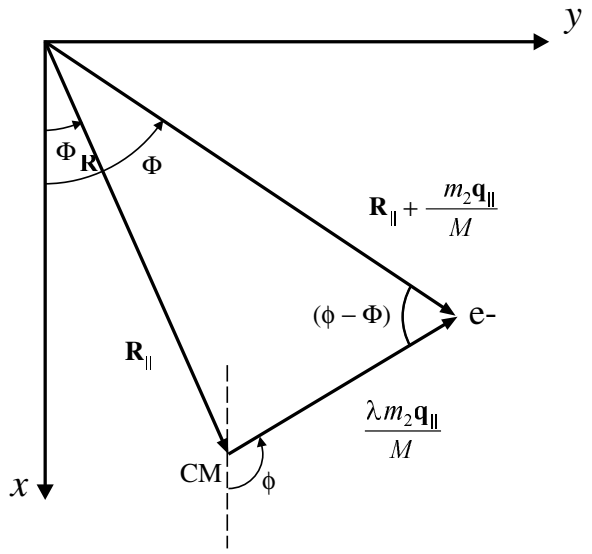

FIG. 2. The vector projections in the x-y plane and the corresponding azimuthal angles for the vectors $\mathbf{R}_\parallel$, $\lambda m_2 \mathbf{q}_\parallel / M$ and their sum $\mathbf{R}_\parallel + \lambda m_2 \mathbf{q}_\parallel / M$ in the context of $H^{(1)}_{\rm int}$.

approximation, the internal motion does not participate in any exchange of momentum with the light beam, neither linear momentum nor orbital angular momentum.

Consider next terms quadratic in vector components of $\mathbf{q}$. These correspond to quadrupole interaction and are classifiable into three kinds. The first is of the form

$$H^{(1)}_{\rm int}(qq) = \alpha \hat{\boldsymbol{\epsilon}} \cdot \mathbf{q} q_z e^{il\Phi_R} F(R_\parallel) e^{ikR_z} e^{-i\omega t} a_{kl} + \text{H.c.}, \tag{22}$$

where α is a constant. The second type is of the form

$$H^{(2)}_{\rm int}(qq) = \beta \hat{\boldsymbol{\epsilon}} \cdot \mathbf{q} q_\parallel e^{i\phi} e^{i(l-1)\Phi_R} G^-_l(R_\parallel) e^{ikR_z} e^{-i\omega t} a_{kl} + \text{H.c.}, \tag{23}$$

where β is a constant. The third is of the form

$$H^{(3)}_{\rm int}(qq) = \beta \hat{\boldsymbol{\epsilon}} \cdot \mathbf{q} q_\parallel e^{-i\phi} e^{i(l+1)\Phi_R} G^+_l(R_\parallel) e^{ikR_z} e^{-i\omega t} a_{kl} + \text{H.c.} \tag{24}$$

It is easy to show that once $H^{(1)}_{\rm int}(qq)$, Eq. (22) is inserted in the matrix element in Eq. (9), this term cannot mediate any transfer of OAM between the light and the internal motion. However, transfer of OAM does occur between the light and the center of mass motion, as in the electric dipole case. By contrast, we see in the expression for $H^{(2)}_{\rm int}(qq)$ in Eq. (23) that a factor $e^{i\phi}$ now appears in the matrix element between internal states $|e\rangle$ and $|g\rangle$, and the center of mass azimuthal phase factor is now $e^{i(l-1)\Phi_R}$. This is indicative of transfer of OAM from the light beam to the internal motion, leaving only $(l-1)\hbar$ which are transferred to the center of mass rotation. Similarly when $H^{(3)}_{\rm int}(qq)$, Eq. (24), is substituted in the matrix element, we conclude that a transfer of OAM occurs between the internal motion and the light beam, enhancing the beam by one unit to $(l+1)\hbar$ which is transferred to the center of mass rotation. It is easy to check that the integrals over the internal azimuthal angle ϕ lead to the usual quadupole selection rule $|m_e - m_g| = 0, \pm 1, \pm 2$, where m_e and m_g are the azimuthal quantum numbers of the respective internal states $|e\rangle$ and $|g\rangle$ involved in the transition.

In conclusion, we have demonstrated by explicit analysis that, in the interaction of light possessing orbital angular momentum with atoms or molecules, the major mechanism of exchange occurs in the electric dipole approximation and involves only the center of mass motion and the light beam. The internal "electronic-type" motion does not participate in any OAM exchange with the light beam to this leading order. It is only in the weaker electric quadrupole interaction that an exchange involving all three subsystems (the light, the atomic center of mass, and the internal motion) can take place. This involves one unit of OAM exchanged between the light beam and the internal motion resulting in the light beam possessing $(l \pm 1)\hbar$, which are then transferred to the center of mass motion. These conclusions rule out any experiments which seek to observe orbital angular momentum exchange involving light beams and the internal states of molecular systems via electric dipole transitions.

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