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Commutation rules and eigenvalues of spin and orbital angular momentum of radiation fields

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Abstract. We investigate the separation of the total angular momentum **J** of the electromagnetic field into a 'spin' part **S** and an 'orbital' part **L**. We show that both 'spin' and 'orbital' angular momentum are observables. However, the transversality of the radiation field affects the commutation relations for the associated quantum operators. This implies that neither **S** nor **L** are angular momentum operators. Moreover their eigenvalues are not discrete. We construct field modes such that each mode excitation (photon) is in a simultaneous eigenstate of S_z and L_z. We consider the interaction of such a photon with an atom and the resulting effect on the internal and external part of the atomic angular momentum.

1. Introduction

As is well known the total mechanical angular momentum **J** of a system of matter particles can be separated into an external and an internal part. One writes

$$\mathbf{J}=\mathbf{L}+\mathbf{S}. \tag{1}$$

The external part **L** gives the angular momentum associated with the centre-of-mass motion. The quantities **L** and **S** obey independent evolution equations, and the corresponding quantum operators commute and both represent observables.

The angular momentum **J** of the free classical electromagnetic field is well defined: it is the conserved quantity resulting from the invariance of the free Maxwell equations under arbitrary rotations [1]. In this case the separation of **J** into spin and orbital angular momentum is known to be impossible [2–5]. There are two main reasons for this. First, the spin part should correspond to the total angular momentum of a particle in its rest frame, but this frame does not exist for a photon. In fact, for a photon one can define only the component of the spin operator along the propagation direction, i.e. the helicity. For a massless spin-s particle the helicity takes the values $\pm s$. Furthermore, the spin is expected to generate rotations of only the polarization of the field, while leaving the amplitude unchanged. This would destroy the transversality of the field, so that the spin operator would not leave the physical subspace invariant [4, 5]. In general, for a massless particle the gauge conditions introduce an interdependence between the vectorial nature and the spatial dependence of the field, and it is not possible to define spin and orbital rotations of the field separately [2].

On the other hand, for the electromagnetic field the vector **J** can be separated into two gauge-invariant parts **L** and **S**, which are often termed the 'orbital' and the 'spin'

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part [4, 5]. The former quantity is similar to external angular momentum, as it is defined relative to a reference point. The 'spin' part is known as the intrinsic part, since it is independent of the choice of this point. For the classical free electromagnetic field both quantities **L** and **S** are conserved. Only in interaction with matter do they change in time (see section 2). The question arises, what is the physical significance of these two quantities **L** and **S**, and how for instance a given component of the 'orbital' angular momentum can be measured.

For light beams, the components of **L** and **S** along the propagation direction can be separately observed by measuring the change of the angular momentum of matter after it has interacted with an appropriate mode of the radiation field. For instance, Beth measured the torque exerted on a birefringent plate, when the plate transforms right circularly polarized light into left circularly polarized light [6]. This has been interpreted as a measurement of the component of the spin angular momentum of light along the propagation (z) direction of the beam. An analogous experiment is being performed in which the z component of the orbital angular momentum of a paraxial laser beam is measured. Here a (monochromatic) Hermite–Gaussian laser mode is transformed into a Laguerre–Gaussian beam by means of two astigmatic lenses [7, 8]. The former mode does not possess angular momentum, whereas the transformed beam has an azimuthal $\exp(im\phi)$ dependence, which implies that the ratio of orbital angular momentum L_z to energy is m/ω, where ω is the frequency of the light. Since L_z is conserved during propagation between the lenses [9], a torque must be exterted on the lens system. In this experiment the polarization state of the light beam and hence S_z are not affected, while in the Beth experiment the spatial field distribution of the light beam and hence L_z are left unchanged. Therefore one is able to measure L_z and S_z *separately*. All these results may be explained by a classical theory.

The physical significance of the quantities **L** and **S** of the radiation field is illustrated by considering the quantized field. It is common practice to quantize the radiation field after expanding it in plane waves. However, two components of the classical angular momentum density in a plane wave diverge for $|\mathbf{r}|\to\infty$. The corresponding two components of **J** are thus ill defined, and so are the components of **L**. Furthermore, the component of the angular momentum density along the wave-vector **k** vanishes identically. This fact leads to paradoxical results (see e.g. [3, 4, 10]): in the Beth experiment the birefringent plate picks up spin angular momentum, while the field apparently does not possess angular momentum. Classically one justifies this result by taking into account the fact that a detector placed in a plane wave causes gradients in this field [4]. The field can no longer be considered as a plane wave, and the z component of the angular momentum density does not vanish. However, in quantum mechanics the problem remains. As is well known, the z component of the internal angular momentum of an atom changes by $\pm\hbar$ on absorption of a photon from a circularly polarized beam moving in that direction. The picture arises that photons in a circularly polarized plane-wave mode possess internal angular momentum $\hbar$ in the propagation direction. This again seems to contradict the fact that J_z vanishes for a plane-wave mode.

In order to investigate the above issues we quantize the field after expanding it into a different complete set of modes. This allows us to quantize the classical quantities **S** and **L**, and determine the commutation relations and eigenvalues for these operators. We compare these with the standard commutation rules and the eigenvalue spectra for spin and orbital angular momenta of fields with spin one. We

find essential differences, which can be traced back to the transversality of the radiation field.

Next, we construct modes for which the photon number states are eigenstates of both L_z and S_z. We subsequently consider the interaction of a photon in such a mode with an atom. One might expect that the 'spin' and 'orbital' angular momentum of the photon are transferred exclusively to, respectively, internal and external angular momentum of the atom. We examine to what extent this statement is correct. We start by considering the classical field in interaction with matter and calculate the concomitant exchange of angular momentum.

2. Angular momentum of the classical field

The electromagnetic field can be separated into transverse and longitudinal fields [1, 5]. These parts have by definition a vanishing divergence and curl, respectively, so that their Fourier transforms are perpendicular and parallel to the vector $\mathbf{k}$ for all $\mathbf{k}$. One uses the symbols $\perp$ and $\parallel$ to denote the transverse and longitudinal parts of the fields. Although this separation is not Lorentz covariant it has nevertheless proved very useful, both in classical theory and for the quantization procedure. The magnetic field is purely transverse, while the longitudinal electric field $\mathbf{E}_{\parallel}$ is given by the instantaneous Coulomb field of the charges. The transverse electric field $\mathbf{E}_{\perp}$ thus describes the radiation part, which contains in fact the only real dynamical degrees of freedom of the field. Moreover, the transverse part $\mathbf{A}_{\perp}$ of the vector potential is gauge invariant.

The angular momentum $\mathbf{J}$ of the classical electromagnetic field relative to the origin is defined by

$$\mathbf{J} = \int \mathrm{d}\mathbf{r}\, \epsilon_0 \mathbf{r} \times (\mathbf{E} \times \mathbf{B}). \tag{2}$$

This quantity is conserved in the absence of charged particles, as a result of the invariance of the free Maxwell equations under spatial rotations. The longitudinal and radiation parts of $\mathbf{E}$ contribute independently to $\mathbf{J}$. The following expressions for the respective contributions can be derived when using $\mathbf{B} = \nabla \times \mathbf{A}_{\perp}$ and by applying partial integration (see also [5], p. 45–47):

$$\begin{aligned} \mathbf{J}_{\mathrm{long}} &= \int \mathrm{d}\mathbf{r}\, \rho(\mathbf{r} \times \mathbf{A}_{\perp}), \\ \mathbf{J}_{\mathrm{rad}} &= \epsilon_0 \sum_i \int \mathrm{d}\mathbf{r}\, E_i^{\perp}(\mathbf{r} \times \nabla) A_i^{\perp} + \epsilon_0 \int \mathrm{d}\mathbf{r}\, \mathbf{E}_{\perp} \times \mathbf{A}_{\perp} \\ &\equiv \mathbf{L}_{\mathrm{rad}} + \mathbf{S}_{\mathrm{rad}}, \end{aligned} \tag{3}$$

with ρ the charge density. It has been assumed here that the fields vanish sufficiently fast for $|\mathbf{r}| \to \infty$ so that surface terms vanish. These expressions are gauge independent, since they do not contain the full vector potential, but only the transverse part $\mathbf{A}_{\perp}$. The longitudinal contribution depends explicitly on the charge density, and is therefore usually combined with the mechanical angular momentum of the (charged and neutral) particles i, which is given by

$$\mathbf{J}_{\mathrm{mech}} = \sum_i m_i \mathbf{r}_i \times \mathbf{v}_i. \tag{4}$$

The sum of mechanical and longitudinal momenta thus gives the momentum associated with the particles i. In the Coulomb gauge this sum is equal to the *canonical* particle angular momentum.

Because of the Lorentz force the mechanical angular momentum changes according to

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{J}_{\mathrm{mech}} = \int \mathrm{d}\mathbf{r}\, \mathbf{r} \times (\rho \mathbf{E}_{\perp} + \mathbf{j} \times \mathbf{B}). \tag{5}$$

The longitudinal part of $\mathbf{E}$ does not contribute here, as ρ is proportional to $\nabla \cdot \mathbf{E}_{\parallel}$. From the Maxwell equations we find the rates of change for the electromagnetic field angular momentum

$$\left.\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}\mathbf{J}_{\mathrm{long}} &= -\int \mathrm{d}\mathbf{r}\, \mathbf{r} \times (\rho \mathbf{E}_{\perp} + \mathbf{j}_{\parallel} \times \mathbf{B}), \\ \frac{\mathrm{d}}{\mathrm{d}t}\mathbf{J}_{\mathrm{rad}} &= -\int \mathrm{d}\mathbf{r}\, \mathbf{r} \times (\mathbf{j}_{\perp} \times \mathbf{B}). \end{aligned}\right\} \tag{6}$$

These relations together with (5) show that the total angular momentum of particles and field is conserved.

Finally we find the time derivatives of $\mathbf{L}_{\mathrm{rad}}$ and $\mathbf{S}_{\mathrm{rad}}$,

$$\left.\begin{aligned} \frac{\partial}{\partial t}\mathbf{L}_{\mathrm{rad}} &= -\sum_i \int \mathrm{d}\mathbf{r}\, j_i^{\perp}(\mathbf{r} \times \nabla) A_i^{\perp}, \\ \frac{\partial}{\partial t}\mathbf{S}_{\mathrm{rad}} &= -\int \mathrm{d}\mathbf{r}\, \mathbf{j}_{\perp} \times \mathbf{A}_{\perp}. \end{aligned}\right\} \tag{7}$$

This result shows that $\mathbf{S}_{\mathrm{rad}}$ and $\mathbf{L}_{\mathrm{rad}}$ are separately conserved in the absence of matter. Furthermore, the change of $\mathbf{S}_{\mathrm{rad}}$ and $\mathbf{L}_{\mathrm{rad}}$ is entirely due to the magnetic interaction with the transverse part $\mathbf{j}_{\perp}$ of the electric current.

3. Angular momentum of the quantized field

3.1. *General mode expansion and quantization*

The radiation field can be expanded in a complete set of transverse mode functions $\mathbf{F}_{\lambda}$, which are solutions of the wave equation

$$\nabla^2 \mathbf{F}_{\lambda} = -k^2 \mathbf{F}_{\lambda}, \tag{8}$$

with the transversality condition

$$\nabla \cdot \mathbf{F}_{\lambda} = 0, \tag{9}$$

and the normalization condition

$$\langle \mathbf{F}_{\lambda} | \mathbf{F}_{\lambda'} \rangle \equiv \int \mathrm{d}\mathbf{r}\, \mathbf{F}_{\lambda}^{*} \cdot \mathbf{F}_{\lambda'} = \delta_{\lambda\lambda'}, \tag{10}$$

where the asterisk denotes complex conjugation. The mode index λ can be specified as the discrete and continuous eigenvalues of mutually commuting Hermitian operators, which have $\mathbf{F}_{\lambda}$ as eigenfunctions. For plane-wave mode functions, the eigenvalues are the three components of the wave-vector $\mathbf{k}$ and the polarization index. In general, mode functions obeying the wave equation (8) are already

eigenfunctions of ∇^2, and three additional independent Hermitian operators must be specified.

The transverse part of the vector potential can be expanded as

$$\mathbf{A}_\perp = \sum_\lambda \mathscr{A}_\lambda (a_\lambda \mathbf{F}_\lambda + a_\lambda^* \mathbf{F}_\lambda^*), \tag{11}$$

with the normalization constant $\mathscr{A}_\lambda = (\hbar/2\epsilon_0\omega_\lambda)^{1/2}$, where $\omega_\lambda = kc$ is the frequency, and with a_λ a dimensionless complex amplitude. With the normalization (10), the energy of the free radiation field takes the form of the energy of a set of independent harmonic oscillators

$$\begin{aligned} H_{\text{rad}} &= \int \mathrm{d}\mathbf{r}\, \epsilon_0 (\mathbf{E}_\perp \cdot \mathbf{E}_\perp + c^2 \mathbf{B} \cdot \mathbf{B})/2 \\ &= \sum_\lambda \frac{\hbar\omega_\lambda}{2} (a_\lambda^* a_\lambda + a_\lambda a_\lambda^*). \end{aligned} \tag{12}$$

Quantization then proceeds in the same way as for harmonic oscillators, where a_λ and $a_\lambda^\dagger = a_\lambda^*$ now become the annihilation and creation operators for photons in the mode λ. They satisfy

$$[a_\lambda, a_{\lambda'}^\dagger] = \delta_{\lambda\lambda'}, \tag{13}$$

while the remaining commutators vanish. The expressions (11) and (12) for $\mathbf{A}_\perp$ and for the field energy are still valid quantum mechanically, provided that $a_\lambda^\dagger$ replaces a_λ^*. The transverse electric and the magnetic field are given by

$$\left.\begin{aligned} \mathbf{E}_\perp &= \sum_\lambda \mathrm{i}\omega_\lambda \mathscr{A}_\lambda (a_\lambda \mathbf{F}_\lambda - a_\lambda^\dagger \mathbf{F}_\lambda^*), \\ \mathbf{B} &= \sum_\lambda \mathscr{A}_\lambda (a_\lambda \nabla \times \mathbf{F}_\lambda + a_\lambda^\dagger \nabla \times \mathbf{F}_\lambda^*). \end{aligned}\right\} \tag{14}$$

These field variables have then become operators. They act, just as a_λ and $a_\lambda^\dagger$, on the Fock space of states of the radiation field. This space is spanned by the photon number states, which can be defined in terms of the vacuum state vector $|0, 0, \ldots\rangle$, according to

$$|\{n_\lambda\}\rangle = \prod_\lambda \frac{(a_\lambda^\dagger)^{n_\lambda}}{(n_\lambda!)^{1/2}} |0, 0, \ldots\rangle, \tag{15}$$

with the vacuum defined by

$$\forall\lambda : a_\lambda^\dagger a_\lambda |0, 0, \ldots\rangle = 0. \tag{16}$$

3.2. *Angular momentum operators*

The operator for the total angular momentum of the transverse field has the same form as for the classical field,

$$\mathbf{J}_{\text{rad}} = \epsilon_0 \int \mathrm{d}\mathbf{r}\, \mathbf{r} \times (\mathbf{E}_\perp \times \mathbf{B}). \tag{17}$$

There is no need to symmetrize this expression, since $\mathbf{E}_\perp$ and $\mathbf{B}$, when evaluated at the same position and time, commute [5]. Just as for the classical field this operator

can be separated into two terms, corresponding to 'spin' and 'orbital' angular momentum. By substituting the expansions (14) one finds

$$\mathbf{J}_{\text{rad}} = \mathbf{L}_{\text{rad}} + \mathbf{S}_{\text{rad}}, \tag{18}$$

where

$$\left.\begin{aligned} \mathbf{S}_{\text{rad}} &= \epsilon_0 \int \mathrm{d}\mathbf{r}\, \mathbf{E}_\perp \times \mathbf{A}_\perp \\ &= \frac{1}{2} \sum_{\lambda\lambda'} (a_\lambda^\dagger a_{\lambda'} + a_{\lambda'} a_\lambda^\dagger) \langle \mathbf{F}_\lambda | \hat{\mathbf{S}} | \mathbf{F}_{\lambda'} \rangle, \\ \mathbf{L}_{\text{rad}} &= \epsilon_0 \sum_l \int \mathrm{d}\mathbf{r}\, E_l^\perp (\mathbf{r} \times \nabla) A_l^\perp \\ &= \frac{1}{2} \sum_{\lambda\lambda'} (a_\lambda^\dagger a_{\lambda'} + a_{\lambda'} a_\lambda^\dagger) \langle \mathbf{F}_\lambda | \hat{\mathbf{L}} | \mathbf{F}_{\lambda'} \rangle. \end{aligned}\right\} \tag{19}$$

Expressions similar to (19) have been derived before both in real space [11] and in reciprocal space [5]. The operators $\hat{\mathbf{L}}$ and $\hat{\mathbf{S}}$ are defined by

$$\left.\begin{aligned} \hat{\mathbf{L}} &= -\mathrm{i}\hbar(\mathbf{r} \times \nabla), \\ (\hat{\mathbf{S}}_k)_{ij} &= -\mathrm{i}\hbar \epsilon_{ijk}, \end{aligned}\right\} \tag{20}$$

for $i,j = x, y, z$, with ϵ_{ijk} the Levi-Civita pseudo tensor. They have the same form as the quantum-mechanical operators for orbital angular momentum and spin of a spin-one particle, respectively. The operator $\hat{\mathbf{L}}$ acts on the position dependence of the classical mode functions $\mathbf{F}_\lambda$, whereas $\hat{\mathbf{S}}$ acts as vector of 3×3 matrices on their cartesian components. We will denote these operators by carets, in order to distinguish them from field operators such as $\mathbf{S}_{\text{rad}}$ and $\mathbf{L}_{\text{rad}}$, which act on Fock states.

It must be noted that the expression for $\mathbf{L}_{\text{rad}}$ may contain additional boundary terms, which result from the integral over all space of a total divergence. Their explicit form may be found in [12]. These boundary terms do not vanish when the mode functions $\mathbf{F}_\lambda$ do not fall off sufficiently fast for $|\mathbf{r}| \to \infty$. This applies in particular to the choice of plane waves as the mode functions. Here we tacitly have assumed that the boundary terms may be neglected. This is correct when, for example, suitable boundary conditions have been imposed, or when a set of square-integrable mode functions has been chosen.

In (19) we already made use of the fact that $\hat{\mathbf{L}}$ and $\hat{\mathbf{S}}$ commute with the Laplace operator, so that only terms with $\omega_\lambda = \omega_{\lambda'}$ contribute to $\mathbf{J}_{\text{rad}}$. Hence, for the free field (19) is manifestly time-independent, and $\mathbf{J}_{\text{rad}}$, $\mathbf{S}_{\text{rad}}$ and $\mathbf{L}_{\text{rad}}$ are conserved, also in the quantum case. Lenstra and Mandel [12] point out that none of the components of $\mathbf{J}_{\text{rad}}$ is strictly conserved for a quantized field with periodic boundary conditions imposed on a cube. The reason is that the quantization volume is not invariant under infinitesimal rotations. On the other hand, $\mathbf{S}_{\text{rad}}$ is always strictly conserved, since the latter operator does not contain boundary terms.

The operators $\hat{\mathbf{S}}$ and $\hat{\mathbf{L}}$, when acting on a transverse field, do not preserve the transversality of that field [5]. If the mode functions $\mathbf{F}_\lambda$ were interpreted as the quantum state vectors for photons, then this would imply that $\hat{\mathbf{S}}$ and $\hat{\mathbf{L}}$ cannot represent observables, since $\hat{\mathbf{S}}|\mathbf{F}_\lambda\rangle$ and $\hat{\mathbf{L}}|\mathbf{F}_\lambda\rangle$ are unphysical states. However, this argument does not apply as the quantum state of the radiation field is described by

Fock states. It is the operators $\mathbf{S}_{\text{rad}}$ and $\mathbf{L}_{\text{rad}}$, and not $\hat{\mathbf{S}}$ and $\hat{\mathbf{L}}$, that act on these Fock states and hence are the physical variables.

Finally, we note that in a similar fashion one may obtain the expression for the linear momentum operator $\mathbf{P}_{\text{rad}}$ of the radiation field,

$$\mathbf{P}_{\text{rad}} = \epsilon_0 \int \mathrm{d}\mathbf{r}\,(\mathbf{E}_\perp \times \mathbf{B})$$

$$= \frac{1}{2} \sum_{\lambda\lambda'} (a_\lambda^\dagger a_{\lambda'} + a_{\lambda'} a_\lambda^\dagger) \langle \mathbf{F}_\lambda | \hat{\mathbf{P}} | \mathbf{F}_{\lambda'} \rangle, \tag{21}$$

with $\hat{\mathbf{P}}$ the quantum-mechanical momentum operator,

$$\hat{\mathbf{P}} = -\mathrm{i}\hbar\nabla. \tag{22}$$

In the remainder of this paper we shall discuss only the momentum $\mathbf{P}_{\text{rad}}$ and the angular momenta $\mathbf{J}_{\text{rad}}$, $\mathbf{L}_{\text{rad}}$ and $\mathbf{S}_{\text{rad}}$ of the radiation field. For convenience we suppress the suffix 'rad' from now on.

3.3. *Commutation relations*

The operators $\hat{\mathbf{S}}$ and $\hat{\mathbf{L}}$ satisfy the standard commutation rules for angular momentum operators

$$[\hat{S}_i, \hat{S}_j] = \sum_k \mathrm{i}\hbar\epsilon_{ijk}\hat{S}_k,$$

$$[\hat{L}_i, \hat{L}_j] = \sum_k \mathrm{i}\hbar\epsilon_{ijk}\hat{L}_k. \tag{23}$$

Since the total angular momentum operator $\mathbf{J}$ generates rotations in space, as was explicitly verified in [12], also $\mathbf{J}$ obeys the standard rule

$$[J_i, J_j] = \sum_k \mathrm{i}\hbar\epsilon_{ijk}J_k. \tag{24}$$

One might expect $\mathbf{S}$ and $\mathbf{L}$ to obey the same relations (23), because of the form (19). However, this turns out not to be true. This is most easily seen if we take the standard expansion of the field in plane-wave modes with wave-vector $\mathbf{k}$, with circular polarization. Then equation (19) reduces to the form obtained before in [12]

$$\mathbf{S} = \sum_{\mathbf{k}} \frac{\hbar\mathbf{k}}{k} (a_{\mathbf{k},+}^\dagger a_{\mathbf{k},+} - a_{\mathbf{k},-}^\dagger a_{\mathbf{k},-}). \tag{25}$$

where the $\pm$ refers to circular polarization with helicity parallel or anti-parallel. Since the operator $\mathbf{S}$ contains only number operators, all its components commute. Therefore we may write

$$[S_i, S_j] = 0. \tag{26}$$

This implies that $\mathbf{S}$ does not generate rotations of the polarization of the field, and it cannot be interpreted as spin angular momentum. Although this result follows directly from (25) it does not seem to have been noticed before.

The commutation rules are, of course, valid in any representation. However, in the plane-wave representation the mode expansion (19) for the operator $\mathbf{L}$ makes no sense. We therefore give now an alternative derivation of the commutation rules for

S and **L**, which will moreover show explicitly that the modification of the standard rules arises from the transversality of the fields.

The expansion (19) for a cartesian component S_i of **S** contains matrix elements of the operator $\hat{S}_i$ between mode functions $\mathbf{F}_\lambda$. These matrix elements do not change their value when the operator $\hat{S}_i$ is replaced by the operator $\hat{T}\hat{S}_i\hat{T}$, where $\hat{T}$ denotes the projection on the space of transverse functions. The action of $\hat{T}$ on the Fourier transform $\mathscr{F}$ of $\mathbf{F}(\mathbf{r})$ is given by the matrix

$$\mathscr{T}_{ij}=\delta_{ij}-\kappa_i\kappa_j, \tag{27}$$

where $\kappa_i=k_i/k$ is the ith component of the unit vector in the **k** direction. In Fourier representation the 'projected' spin operator takes the form

$$(\mathscr{T}\hat{S}_k\mathscr{T})_{ij}=-\mathrm{i}\hbar[\epsilon_{ijk}-\sum_s(\epsilon_{isk}\kappa_s\kappa_j-\epsilon_{jsk}\kappa_s\kappa_i)]. \tag{28}$$

This operator is a vector operator of antisymmetric 3×3 matrices. These matrices are transverse in the sense that when acting on κ_i they give a vanishing result. Now there is only a single antisymmetric transverse matrix, namely the matrix with components $\Sigma_s\epsilon_{ijs}\kappa_s$. Hence each component of the operator $\mathscr{T}\hat{\mathbf{S}}\mathscr{T}$ is proportional to this very matrix. In fact, one finds

$$(\mathscr{T}\hat{S}_k\mathscr{T})_{ij}=-\mathrm{i}\hbar\sum_s\epsilon_{ijs}\kappa_s\kappa_k, \tag{29}$$

which can be verified by taking the inner product of (28) and (29) with the vector **k**. All the components of $\hat{T}\hat{\mathbf{S}}\hat{T}$ therefore must commute. Therefore the mode functions $\mathbf{F}_\lambda$ can be chosen as simultaneous eigenfunctions of $\hat{T}\hat{S}_x\hat{T}$, $\hat{T}\hat{S}_y\hat{T}$ and $\hat{T}\hat{S}_z\hat{T}$. In such a representation, the double summation in (19) reduces to a single summation, and all components of **S** depend exclusively on number operators $a_\lambda^\dagger a_\lambda$. Since all number operators are mutually commuting, we conclude that the components of **S** are commuting operators on the Fock space of state vectors of the radiation field.

In fact, the commutation rule (26) can also be proven by starting from the definition (19) of **S**, and applying the commutation relation (see [5], p. 173)

$$[A_{\perp,i}(\mathbf{r}),E_{\perp,j}(\mathbf{r}')]=\frac{\hbar}{\mathrm{i}\epsilon_0}\delta_{ij}^{\perp}(\mathbf{r}-\mathbf{r}'), \tag{30}$$

with $\delta_{ij}^{\perp}(\mathbf{r}-\mathbf{r}')$ the transverse delta function.

The commutation relations between **J** and **S** read

$$[J_i,S_j]=\sum_k\mathrm{i}\hbar\epsilon_{ijk}S_k, \tag{31}$$

which shows that **S** transforms as a vector under rotations. The same is then true for **L**. From the three relations (24), (26) and (31) it follows that the 'orbital angular momentum operator' **L** obeys

$$\left.\begin{aligned}[L_i,L_j]&=\sum_k\mathrm{i}\hbar\epsilon_{ijk}(L_k+S_k),\\ [L_i,S_j]&=\sum_k\mathrm{i}\hbar\epsilon_{ijk}S_k.\end{aligned}\right\} \tag{32}$$

The first line implies that also **L** is not an angular momentum operator. It does not generate orbital rotations. The second line shows that **L** and **S** do not commute,

unlike $\hat{\mathbf{L}}$ and $\hat{\mathbf{S}}$, and unlike the corresponding operators for internal and external angular momentum of matter particles. On the other hand the operator $\mathbf{L}$ does commute with $\mathbf{S}^2$. Also, it is clear from the form (25) of the spin operator that $\mathbf{S}$ and the linear momentum operator $\mathbf{P}$ can be simultaneously diagonalized. Therefore these two field operators commute. Since the total angular momentum generates rotations of the orbital vector $\mathbf{P}$ it follows subsequently that

$$[L_i, P_j] = \sum_k i\hbar\epsilon_{ijk}P_k. \tag{33}$$

Finally, we know that the 'spin' operator $\mathbf{S}$ cannot generate general rotations of the polarization. Instead it generates transformations of the polarization vector such that the transversality of the field is preserved. From the form (25) it becomes clear that for an arbitrary unit vector $\mathbf{u}$, the operator

$$R(\alpha) = \exp(-i\alpha\mathbf{u}\cdot\mathbf{S}/\hbar) \tag{34}$$

rotates the polarization of each $\mathbf{k}$-component of the field around its wave-vector $\mathbf{k}$, over an angle $\alpha\cos(\mathbf{u}\cdot\mathbf{k}/k)$. Hence the effective rotation vector is the projection of $\mathbf{u}$ along the wave-vector $\mathbf{k}$. This shows once more that only the component of the operator $\mathbf{S}$ along $\mathbf{k}$ is a true spin angular momentum operator, since only this component generates spin rotations.

4. Eigenstates of spin and orbital angular momentum

In order to study the transfer of $\mathbf{L}$ and $\mathbf{S}$ to an atom at absorption of a photon, we wish to construct a set of transverse modes in such a way that each photon from a given mode will be in an eigenstate of both the spin and the orbital angular momentum in a given (z) direction. Then the operators S_z and L_z are given by expansions (19) which are diagonal in λ.

We follow a method of construction which is similar to the method used by Berestetskii to construct the familiar multipolar waves [2]. We start by defining a complete set of scalar functions f which solve the wave equation. We define the functions f by requiring them to be eigenfunctions of three commuting hermitian operators. Then this set of scalar functions is complete and orthogonal. We then proceed to construct from these scalar functions transverse vector eigenfunctions of the operators $\hat{\mathbf{P}}^2$, $\hat{P}_z$ and of $\hat{J}_z$. There are two independent solutions. Subsequently we find the two linear combinations of these solutions which are eigenfunctions of the reduced spin operator $\hat{T}\hat{S}_z\hat{T}$. These modes describe then eigenstates of energy and of the field operators J_z, P_z and S_z. Note that the multipolar waves correspond to eigenstates of energy, J_z, $\mathbf{J}^2$ and parity.

4.1. *Scalar functions*

We introduce a complete set of scalar eigenfunctions f of the commuting operators $\hat{L}_z$, $\hat{\mathbf{P}}^2$ and $\hat{P}_z$, so that

$$\left.\begin{aligned}(\hat{P}_x^2 + \hat{P}_y^2)f &= \hbar^2 k_t^2 f,\\ \hat{P}_z f &= \hbar k_z f,\\ \hat{L}_z f &= m\hbar f,\end{aligned}\right\} \tag{35}$$

where $k_t^2 = k^2 - k_z^2$. We denote the so-constructed functions by $f(k_t, k_z, m)$. They can be written in cylindrical coordinates as

$$f(k_t, k_z, m) = J_m(k_t\rho)\exp(ik_z z)\exp(im\phi)/N, \tag{36}$$

with $J_m(k_t\rho)$ a Bessel function and N a normalization constant. We may normalize them according to

$$\int \mathrm{d}\mathbf{r}\, f^*(k_t, k_z, m) f(k_t', k_z', m') = \delta(k_z - k_z')\delta(k_t - k_t')\delta_{mm'}. \tag{37}$$

4.2. *Mode functions*

Each orbital vector operator $\hat{\mathbf{V}}$ satisfies the commutation relation

$$[\hat{L}_z, \hat{V}_j] = \sum_k i\hbar\epsilon_{zjk}\hat{V}_k. \tag{38}$$

This relation implies that a vector eigenfunction of $\hat{J}_z = \hat{L}_z + \hat{S}_z$ can be constructed by letting an arbitrary orbital vector operator act on a scalar eigenfunction of the operator L_z. This statement remains true when the 'vector' operator transforms as a vector only with respect to O(2), under rotations about the z axis, which is sufficient for the validity of (38). Since there are two independent choices for selecting vectors perpendicular to $\mathbf{k}$ in Fourier space we wish to find two commuting vector operators $\hat{\mathbf{V}}_1$ and $\hat{\mathbf{V}}_2$ such that the set of functions

$$\mathbf{F}(k_t, k_z, m, s) = \frac{\hat{\mathbf{V}}_s f(k_t, k_z, m)}{(\langle \hat{\mathbf{V}}_s f | \hat{\mathbf{V}}_s f \rangle)^{1/2}}, \tag{39}$$

for $s = 1, 2$ forms a complete set of normalized transverse vector eigenfunctions of $\hat{\mathbf{P}}^2$, $\hat{P}_z$ and $\hat{J}_z$. We impose the following conditions on $\hat{\mathbf{V}}_s$

$$\left.\begin{aligned} \hat{\mathbf{P}}\cdot\hat{\mathbf{V}}_s = 0, \\ \hat{\mathbf{V}}_1^\dagger\cdot\hat{\mathbf{V}}_2 = 0, \end{aligned}\right\} \tag{40}$$

with the dagger denoting the hermitian conjugate. The first condition ensures transversality, the second orthogonality of the mode functions. Furthermore, we require these operators to commute with $\hat{\mathbf{P}}^2$ and with $\hat{P}_z$, so that the vector functions (39) still satisfy the first two eigenvalue equations (35). Then the modes will possess well-defined energy and linear momentum in the z direction. The most obvious choice is to take

$$\hat{\mathbf{V}}_1 = \hat{\mathbf{P}} \times \hat{\mathbf{z}}, \qquad \hat{\mathbf{V}}_2 = \hat{\mathbf{V}}_1 \times \hat{\mathbf{P}}. \tag{41}$$

The first operator defines modes with a vanishing E_z, i.e. TE modes. The modes with $s = 2$ are TM modes, since $B_z = 0$.

Finally we have to find two linear combinations $\hat{\mathbf{V}}_\pm$ of $\hat{\mathbf{V}}_1$ and $\hat{\mathbf{V}}_2$ such that $\hat{\mathbf{V}}_+ f$ and $\hat{\mathbf{V}}_- f$ are eigenvectors of the reduced spin operator $\hat{T}S_z\hat{T}$,

$$(\hat{T}\hat{S}_z\hat{T})\hat{\mathbf{V}}_\pm f = \mu_\pm \hat{\mathbf{V}}_\pm f, \tag{42}$$

with eigenvalues $\mu_\pm$. Then the modes $\hat{\mathbf{V}}_\pm f$ will possess well-defined spin in the z direction. From (29) it is obvious that in Fourier space, the two vectors $\hat{\mathbf{V}}_+$ and $\hat{\mathbf{V}}_-$ obeying the requirements (40) and (42) are the two circular vectors in the plane normal to $\mathbf{k}$. They are eigenvectors of $\hat{T}\hat{S}_z\hat{T}$ with eigenvalues $\pm\hbar\kappa_z$. The

corresponding expressions for the vector operators acting on the scalar functions f are

$$\hat{\mathbf{V}}_{\pm}=\frac{1}{\sqrt{2}}\left(\frac{\hat{\mathbf{V}}_2}{\hbar^2 k k_t}\pm\frac{\mathrm{i}\hat{\mathbf{V}}_1}{\hbar k_t}\right), \tag{43}$$

where we included the proper normalization factors, which follow directly from

$$\left.\begin{aligned}\hat{\mathbf{V}}_1^2&=\hat{P}_x^2+\hat{P}_y^2,\\ \hat{\mathbf{V}}_2^2&=\hat{P}^2(\hat{P}_x^2+\hat{P}_y^2).\end{aligned}\right\} \tag{44}$$

The corresponding mode functions

$$\mathbf{F}(k_t, k_z, m, \pm)=\hat{\mathbf{V}}_{\pm}f(k_t, k_z, m) \tag{45}$$

are normalized according to

$$\int \mathrm{d}\mathbf{r}\,\mathbf{F}^*(k_t, k_z, m, s)\cdot\mathbf{F}(k_t', k_z', m', s')=\delta(k_z-k_z')\delta(k_t-k_t')\delta_{mm'}\delta_{ss'}. \tag{46}$$

Their cartesian components are given by†

$$\left.\begin{aligned}\frac{1}{\sqrt{2}}(F_x+\mathrm{i}F_y)&=\frac{k_z\mp k}{2k}f(k_t, k_z, m+1),\\ \frac{1}{\sqrt{2}}(F_x-\mathrm{i}F_y)&=\frac{k_z\pm k}{2k}f(k_t, k_z, m-1),\\ F_z&=\frac{k_t}{\sqrt{2}k}f(k_t, k_z, m).\end{aligned}\right\} \tag{47}$$

These mode functions are eigenfunctions of the operators $\hat{\mathbf{P}}^2$, P_z, $\hat{J}_z$ and $\hat{T}\hat{S}_z\hat{T}$ with eigenvalues $\hbar^2k^2$, $\hbar k_z$, $m\hbar$ and $\pm\hbar k_z/k$.

The mode functions (47) are in fact generalizations of so-called Bessel beams, which have recently been experimentally realized [13]. Bessel beams are proportional to the zeroth-order Bessel function $J_0(k_t\rho)$, and do not depend on the azimuthal angle ϕ. Our modes, on the other hand, carry arbitrary orbital angular momentum. They belong to the class of non-diffracting beams [14], since the intensity profile is independent of the propagation variable z.

Finally, in relation to the remarks made after equation (19) on the neglect of boundary terms, we note that the mode functions $f(k_t, k_z, m)$ are not square integrable, and do not vanish for $z\to\infty$. The Bessel functions do approach zero as

$$J_m(k_t\rho)\sim\left(\frac{2}{\pi k_t\rho}\right)^{1/2}\cos(k_t\rho-\tfrac{1}{2}m\pi-\tfrac{1}{4}\pi), \tag{48}$$

for $\rho\to\infty$, but this is too slow to make the boundary terms in $\mathbf{J}-\mathbf{S}$ vanish. However, the z component L_z is still given by (19) if suitable boundary conditions are imposed on a cylinder $\rho=$constant. Moreover, since this quantization volume is invariant under rotations about the z axis, L_z and J_z are strictly conserved in time.

† We define the relative phases of the functions $f(m)$ for different m by $(\hat{P}_x\pm\mathrm{i}\hat{P}_y)f(m)=-\hbar k_t f(m\pm1)$.

4.3. *Angular momentum operators*

The modes $\lambda \equiv (k_t, k_z, m, s)$ just defined have well-defined values for the angular momentum J_z of $m\hbar$ per photon. More precisely, on substituting the expansions (14) for $\mathbf{E}_\perp$ and $\mathbf{B}$ one finds

$$J_z = \int \mathrm{d}k_t \int \mathrm{d}k_z \sum_{m,s} m\hbar \hat{N}_\lambda, \tag{49}$$

in terms of the number operators $\hat{N}_\lambda = a_\lambda^\dagger a_\lambda$. By construction the operator S_z is diagonal in the eigenstates of these number operators

$$S_z = \int \mathrm{d}k_t \int \mathrm{d}k_z \sum_{m,s} \frac{s\hbar k_z}{k} \hat{N}_\lambda. \tag{50}$$

The eigenvalues of this operator are given by $\pm k_z\hbar/k$ per photon. This follows immediately from the eigenvalues of the projected spin operator. This result confirms that S_z is not a true angular momentum operator, since then its eigenvalues would have been discrete, viz. $\pm\hbar$ per photon. The operator for orbital angular momentum L_z is

$$L_z = \int \mathrm{d}k_t \int \mathrm{d}k_z \sum_{m,s} \left(m\hbar - \frac{s\hbar k_z}{k} \right) \hat{N}_\lambda. \tag{51}$$

Again, the eigenvalues $m\hbar \mp k_z\hbar/k$ per photon of this operator are not discrete.

5. Interaction with atoms

The creation operators $a_\lambda^\dagger$ acting on the vacuum state result in state vectors of the radiation field that are eigenvector of the total angular momentum in the z direction with eigenvalue $m\hbar$, of the spin part $S_z(\pm k_z\hbar/k)$, and hence also of the orbital part L_z; furthermore of the z component of the linear momentum $P_z(\hbar k_z)$, and of the energy $H_{\mathrm{rad}}(\hbar kc)$.

We now want to investigate where the quantities L_z and S_z go when a photon is absorbed by an atom. The total angular momentum of the atom is separated as

$$\mathbf{J}_{\mathrm{at}} = \mathbf{J}_{\mathrm{ext}} + \mathbf{J}_{\mathrm{int}}, \tag{52}$$

where

$$\mathbf{J}_{\mathrm{ext}} = \mathbf{R} \times \mathbf{P}, \tag{53}$$

with $\mathbf{R}$ the position of the centre of mass, and $\mathbf{P}$ the total momentum of the atom, and where $\mathbf{J}_{\mathrm{int}}$ is the total internal angular momentum. The atom–field interaction in the dipole approximation is described by the interaction Hamiltonian

$$H_{\mathrm{I}} = -\mathbf{d} \cdot \mathbf{E}(\mathbf{R}). \tag{54}$$

It must be noted that this approximation does not neglect the spatial dependence of the field, so that the orbital angular momentum of the field is still included. The dipole operator can be written in terms of internal atomic operators alone, and external operators occur only through $\mathbf{E}$, which is given by an expansion of the form (14). As a consequence, in a matrix element $\langle f|H_{\mathrm{I}}|i\rangle$ between some initial and some final state, the position dependence of the field mode determines the selection rules concerning external variables, whereas the vectorial character determines the selection rules for internal quantities.

5.1. *Transfer of angular momentum*

We consider an initial state $|M_{\text{ext}}, M_{\text{int}}; \lambda\rangle$, where a single atom is in simultaneous eigenstates of the z components of external and internal atomic angular momentum with eigenvalues M_{ext} and M_{int} in units $\hbar$, where the external momentum is defined relative to the origin of the coordinate system. The remaining atomic quantum numbers are left unspecified. The radiation field contains one photon in the mode λ defined above, i.e. in an eigenstate of the field operators S_z and L_z.

This state is coupled, by absorption of the photon, to a final state $|f\rangle$ of the form

$$|f\rangle = \sum_{M'_{ext}, M'_{int}} \mathscr{A}_{M'_{ext}, M'_{int}} |M'_{\text{ext}}, M'_{\text{int}}; 0\rangle, \tag{55}$$

with zero photons. The amplitudes $\mathscr{A}$ are proportional to the matrix elements

$$\langle M'_{\text{int}}, M'_{\text{ext}}; 0 | \omega_\lambda \mathscr{A}_\lambda \mathbf{d} \cdot \mathbf{F}_\lambda(\mathbf{R}) a_\lambda | M_{\text{int}}, M_{\text{ext}}; \lambda \rangle \tag{56}$$

of the appropriate part of the interaction Hamiltonian H_1.

Now the field $\mathbf{F}_\lambda$ is given by (47). This implies that the final atomic state is in general no longer an eigenstate of internal and external angular momentum, but consists of a superposition of three different kinds of states. Namely, the angular momentum J_z of the photon, $m\hbar$, can be divided among internal and external degrees of freedom of the atom in three ways: the change in external angular momentum δM_{ext} of the atom may be equal to $\delta M_{\text{ext}} = (m-1), m, (m+1)$. This follows from the matrix elements†

$$\left.\begin{aligned}
\langle M'_{\text{int}}, M'_{\text{ext}} | (F_x - \mathrm{i}F_y)(x + \mathrm{i}y) | M_{\text{int}}, M_{\text{ext}} \rangle &\propto \delta[M'_{\text{ext}} - M_{\text{ext}} - (m-1)] \\
&\quad \times \delta(M'_{\text{int}} - M_{\text{int}} - 1), \\
\langle M'_{\text{int}}, M'_{\text{ext}} | (F_x + \mathrm{i}F_y)(x - \mathrm{i}y) | M_{\text{int}}, M_{\text{ext}} \rangle &\propto \delta[M'_{\text{ext}} - M_{\text{ext}} - (m+1)] \\
&\quad \times \delta(M'_{\text{int}} - M_{\text{int}} + 1), \\
\langle M'_{\text{int}}, M'_{\text{ext}} | F_z z |, M_{\text{int}}, M_{\text{ext}} \rangle &\propto \delta(M'_{\text{ext}} - M_{\text{ext}} - m) \\
&\quad \times \delta(M'_{\text{int}} - M_{\text{int}}).
\end{aligned}\right\} \tag{57}$$

The concomitant change in the internal angular momentum is given by $\delta M_{\text{int}} = 1, 0, -1$, respectively, so that total angular momentum is, indeed, conserved. The relative transition probabilities corresponding to $\delta M_{\text{int}} = 1, 0, -1$ are determined by the corresponding Clebsch–Gordan coefficient and by the integral over the atomic external wavefunctions of the appropriate spherical component of the field mode, as given in (47).

Only for a mode with $k_t = 0$, i.e. $k_z = \pm k$, is it true that the 'spin' and 'orbital' angular momentum of a photon are converted into internal and external angular momentum of an atom, respectively. In that case only one of the spherical components is different from zero, and S_z and L_z can be detected in a single measurement of the change in angular momentum of the atom. Perhaps not surprisingly, such a mode corresponds to a circularly polarized *plane wave* propagating in the z direction. However, the case $k_t = 0$ is singular, in that only the modes with vanishing orbital angular momentum (and $S_z = \pm\hbar$) survive, since for all $m \neq 0$ the Bessel functions $J_m(k_t\rho)$ vanish everywhere for $k_t = 0$. It also shows that the

† A matrix element $\langle f|H_1|i\rangle$ consists of the sum of three factorized terms, as the internal and external atomic operators and the photon operators commute.

angular momentum along the propagation direction of a photon in a plane wave does not vanish. This contradicts the fact that the corresponding component of the classical angular momentum density vanishes in a plane wave. The difference is due to the erroneous neglect of boundary terms in the transition from (17) to (19). However, since this latter result is consistent with angular momentum conservation, as just shown, it seems that the expression (19) is to be preferred over the expression (17).

Also in a circularly polarized paraxial Laguerre–Gaussian beam, it is well possible to measure the orbital angular momentum of one photon [7–9]. Such a beam is a superposition of coherent states of modes $(k_t, k_z, m, \pm)$ with $k_t \ll k$. This latter condition implies that only one of the components (47) is appreciable, while the other two are negligibly small. Hence in a paraxial beam, the quantity $S_z + J_{\text{int},z}$ is separately conserved, as is $L_z + J_{\text{ext},z}$. An important distinction with plane waves is, that now modes with arbitrary integer values for L_z exist.

The atomic states considered so far had well-defined values for angular momentum M_{ext} relative to the origin. This implies that the centre-of-mass part of the atomic wavefunction is centred around the z axis. On the other hand, if an atom has a well-defined position outside this axis, then the atom must be in a superposition of states with different M_{ext}. Now consider an atom placed outside the centre of a Laguerre–Gaussian laser beam with azimuthal index m. When it absorbs a laser photon, it will end up in the same superposition but with each M_{ext} shifted by m. This reflects the conservation law for external angular momentum valid for paraxial beams.

6. Conclusions

We considered the angular momentum **J** of electromagnetic radiation, and its standard separation in the external (orbital) and intrinsic (spin) part. It is generally believed that this separation is unphysical. However, we demonstrate that for the quantized radiation field, the operators **L** and **S** are both Hermitian and gauge-invariant, which implies that both operators represent observable quantities. Furthermore, both for classical and quantum radiation fiels, both quantities vary only due to their interaction with the transverse part of the currents. Therefore, for the free radiation field, **L** and **S** are separately conserved, as is the field momentum **P**.

For the quantized field, the operators **L**, **S** and **P** can be expressed as a summation of matrix elements of operators acting on the mode functions in which the field is expanded. These operators, indicated as $\hat{\mathbf{L}}$, $\hat{\mathbf{S}}$ and $\hat{\mathbf{P}}$ have the same form as the standard quantum-mechanical operators for spin, orbital angular momentum and momentum of a particle with spin one. We stress that these operators do not represent observable quantities of the radiation field. The physical meaning of the operators **L** and **S** is further clarified by determining their commutation rules. We notice that the three components of **S** commute, and that they have non-vanishing commutators with the components of **L**. Hence, since these operators do not obey the commutation rules for the components of angular momentum, they do not generate rotations, and they do not represent proper angular momenta. On the other hand, the components of the total angular momentum **J** do obey the correct commutation rules. The transformation generated by the component S_z of **S** represents a rotation of the polarization of each plane wave mode about the propagation direction, over an angle that is reduced by the cosine of the angle between this propagation direction and the z axis.

Finally, we investigated what happens to the separate parts **S** and **L** of the field angular momentum when a photon is absorbed by an atom. For this purpose we constructed a complete set of transverse mode functions, such that photons in these modes have well-defined values for L_z, S_z, P_z, and energy. There is no conservation law for intrinsic and external angular momentum separately, such as does exist for total angular momentum, or for total linear momentum. In fact, this would be impossible, since the eigenvalues in general no longer match: M_{int}, M_{ext} are discrete, S_z, L_z are continuous. Separate conservation laws do exist for paraxial beams running in the z direction. Moreover, an atom initially in an eigenstate of internal (external) angular momentum will in general not remain in such a state after absorption of a photon in an eigenstate of $S_z(L_z)$, except, again, for the paraxial beam in the z direction.

The conclusion is that both 'spin' **S** and 'orbital' angular momentum **L** of a photon are well defined and separately measurable. This concerns all three components. However, only the components along the propagation direction can be measured by detecting the change in internal and external angular momentum of an atom, respectively.

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