ANGULAR MOMENTUM OF PHOTONS EMITTED BY ATOMS

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It is shown that the spin and orbital angular momentum of electric dipole photons have the same operator structure and may differ from each other only by spatial dependence in the very vicinity of the atom. It is shown that the photon twins created by a dipole forbidden transition can manifest the maximum entanglement with respect to the angular momentum. It is shown that the states of photons with projection of angular momentum $m = 0$ are less stable than those with $m = \pm 1$.

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Recently the problem of angular momentum (AM) of photons has attracted a great deal of interest (e.g., see [1, 2, 3, 4, 5, 6, 7, 8] and references therein), especially in the context of quantum information processing [3, 4, 5, 6, 7, 8, 9]. Usually AM of photons is considered as consisting of the *spin* and *orbital* angular momentum (OAM) parts [10]

$$\vec{M} = \frac{1}{4\pi c}\int \vec{r}\times(\vec{E}\times\vec{B})d^3r = \vec{S}+\vec{L}, \qquad (1)$$

$$\vec{S} = \frac{1}{4\pi c}\int (\vec{E}\times\vec{A})d^3r, \qquad (2)$$

$$\vec{L} = \frac{1}{4\pi c}\sum_{\alpha=x,y,z}\int E_\alpha(i\vec{\mathcal{M}})A_\alpha d^3r. \qquad (3)$$

Here $\vec{\mathcal{M}} \equiv -i\vec{r}\times\vec{\nabla}$ is the quantum mechanical OAM operator and $\vec{E}, \vec{B}, \vec{A}$ denote the electric field, magnetic induction, and vector potential. Within the quantum domain, these objects are represented by linear forms in photon operators of creation and annihilation, so that (1), (2), and (3) are quadratic forms with respect to photon operators. In this case, the integrands in (1)-(3), representing the densities of the corresponding quantities, are the operators defined in the real space $\mathbb{R}^3$ and Hilbert-Fock space of photons.

It is known that a photon has spin 1, but can be observed in only two spin states (helicities) because of the requirement of Pincaré invariance on the light cone [11]. This means that the AM operator (1) is specified by the dynamic symmetry group $G = E(2)\times SU(2)$. Here $E(2)$, describing the photon spin symmetry, is Winger's little group, that is the Euclidean group, which is a semi-direct product of $SO(2)$ and $T(2)$ - the group of translations in two dimensions. In turn, $SU(2)$ in G describes the symmetry of OAM.

Nevertheless, consideration of specific models can lead to a different results. For example, a monochromatic plane wave (both classical and quantum), travelling in the z-direction, has no AM about the z-axis because the Poynting vector $\vec{E}\times\vec{B}$ is parallel to this axis. At the same time, it has spin (2) directed along the z-axis. The rejection of monochromaticity leads to the following commutation relations [12]

$$[S_\alpha, S_\beta] = 0, \quad [L_\alpha, L_\beta] = i\epsilon_{\alpha\beta\gamma}(L_\gamma + S_\gamma),$$
$$[L_\alpha, S_\beta] = i\epsilon_{\alpha\beta\gamma}S_\gamma, \qquad (4)$$

corresponding to even more degenerated group $G' = T(3)\times SU(2)$.

The aim of this work is to examine the structure of the total AM and its spin and OAM parts in the representation of *spherical waves* that corresponds to real photons are emitted by the atomic and molecular transitions [13, 14]. The wave functions of spherical photons are represented by vector spherical harmonics, which are the linear combinations of ordinary spherical harmonics (eigenstates of OAM operator) and spin states [13]. This assumes the $SU(2)\times SU(2)$ structure of the total AM of a photon.

We consider the densities of spin and OAM operators in the representation of spherical (multipole) photons (integrands in (2) and (3)). We show that these operators have the same structure in terms of photon operators but different spatial dependence at short and intermediate distances. In the wave (far) zone, this difference vanishes, so that there is no way to distinguish between the spin and OAM parts of the total AM of a multipole photon. Besides that, we consider generation of photon twins entangled with respect to the total angular momentum.

As an example of some considerable interest, let us investigate an electric dipole ($E1$) transition between the excited $|e;m\rangle$ and ground $|g\rangle$ states with the AM $j_e = 1$ and $j_g = 0$, respectively. Since the angular momentum $j_e = 1$ has the three projections $m = 0, \pm 1$, the excited atomic state $|e;m\rangle$ is triple degenerated. Assume first that such a two-level atom is located at the center of an ideal spherical cavity of high radius R (the multipole Jaynes-Cummings model [15]). Then, the transition

$$|e;m\rangle \to |g\rangle$$

gives rise to a monochromatic $E1$ photon with projection m of AM. The operator vector potential, describing this

photon, has the form [13, 14]

$$\vec{A}_k(\vec{r}) = \sum_m \vec{\mathcal{A}}_{km}(\vec{r}) a_{km} + H.c.$$

$$= \sum_m N(k)[j_2(kr)\vec{Y}_{1,2,m} - \sqrt{2} j_0(kr)\vec{Y}_{1,0,m}]a_{km} + H.c., \quad (5)$$

where $\vec{Y}_{j\ell m}$ is the vector spherical harmonics [13], $N(k) = \sqrt{4\pi\hbar c/(3kV)}$ is the normalization factor, $V = 4\pi R^3/3$ denotes the cavity volume, $j_\ell(kr)$ is a function proportional to the spherical Bessel function but normalized by the condition

$$\int_0^R j_\ell(kr) j_\ell(k'r) r^2 dr = V\delta_{kk'}, \quad (6)$$

and a_{km} are the photon operators

$$[a_{km}, a^+_{k'm'}] = \delta_{kk'}\delta_{mm'}. \quad (7)$$

Taking into account that $\vec{E} = -\partial\vec{A}/(c\partial t)$, we can conclude that the integrands in (2) and (3) contain the operator constructions of the form $a^+_m a_{m'}$. This means that, in contrast to energy, there is no vacuum oscillations of AM.

To specify the spin and OAM at any distance r from the atom, we take into account that the photon localization appears in the natural way in the form of wavefront [16] (concerning photon localization also see [9, 17, 18] and references therein). Therefore, to find AM carried by a photon at distance r from the atom, we should perform integration over the spherical shall of radius r, surrounding the source. Finally, we get

$$\vec{S}(r) = f_S(kr)\vec{J}, \quad \vec{L}(r) = f_L(kr)\vec{J}. \quad (8)$$

Here $\vec{J}$ denotes the operator of the total AM in the whole volume of quantization, having the components

$$\begin{cases} J_x = [a^+_{k0}(a_{k+} + a_{k-}) + H.c.]/\sqrt{2} \\ J_y = i[a^+_{k0}(a_{k+} - a_{k-}) - H.c.]/\sqrt{2} \\ J_z = a^+_{k+}a_{k+} - a^+_{k-}a_{k-} \end{cases} \quad (9)$$

so that

$$[J_\alpha, J_\beta] = i\epsilon_{\alpha\beta\gamma}J_\gamma. \quad (10)$$

Thus, Eqs. (9) give a representation of the $SU(2)$ subalgebra in the Weyl-Heisenberg algebra of photon operators (7).

The distance-dependent functions in (8) have the form

$$f_S(kr) = \frac{\hbar}{3V}\left[2j_0^2(kr) - \frac{1}{2}j_2^2(kr)\right],$$

$$f_L(kr) = = \frac{\hbar}{3V}\frac{3}{2}j_2^2(kr).$$

It follows from the normalization condition (6) that

$$\int_0^R f_S(kr) r^2 dr = \int_0^R f_L(kr) r^2 dr = \frac{\hbar}{2},$$

so that the total AM operator (1) takes the form

$$\vec{M} = \hbar\vec{J}$$

as all one can expect for $E1$ photon with AM equal to one. The total spin and OAM operators have the form

$$\vec{S} = \vec{L} = \frac{\hbar}{2}\vec{J}.$$

The structure (9) of the AM operator can also be obtained in a different way through the use of conservation of the total AM in the process of atom-photon interaction [1, 9].

Thus, spin and OAM of $E1$ photon have the same operator structure. In particular, this means that the commutation relations for the components of the density operators in (2) and (3) take the form

$$\begin{aligned} [S_\alpha(r), S_\beta(r)] &= i\epsilon_{\alpha\beta\gamma} f_S(kr) S_\gamma(r), \\ [L_\alpha(r), L_\beta(r)] &= i\epsilon_{\alpha\beta\gamma} f_L(kr) L_\gamma(r), \\ [L_\alpha(r), S_\beta(r)] &= i\epsilon_{\alpha\beta\gamma} f_L(kr) S_\gamma(r), \end{aligned} \quad (11)$$

in contrast to (4) and (5).

It is seen that the spin and OAM density operators have different spatial dependence at short distances from the atom. Since

$$\lim_{x\to 0} j_\ell(x) = \begin{cases} 1, & \text{if } \ell = 0 \\ 0, & \text{otherwise} \end{cases}$$

$f_L(kr)$ vanishes at $kr \to 0$. Thus, at the very vicinity of the atom, the photon has only spin, while OAM arises in the process of propagation. A more detailed investigation shows that spin density strongly prevails over OAM density at $r < 0.1\lambda$, where $\lambda = 2\pi/k$ is the wavelength. Since the maximum of $f_S(kr)$ corresponds to $kr = 0$, it is possible to say that atom creates the photon with spin and without OAM. In turn, OAM achieves maximum at $r \sim \lambda/2$ (intermediate zone). It is also seen that the main contribution into the total AM comes from the near zone in contrast to the energy that derives its main contribution from the wave zone [19].

Let us stress that the results for monochromatic multipole photons in the near and intermediate zones should be taken carefully. The point is that any excited atomic state has a finite life time (even in a cavity) and therefore the radiation is specified by a certain line width that should be taken into account.

At far distances, we have

$$j_\ell \sim \frac{1}{x}\sin(x - \ell\pi/2), \quad x = kr \gg \ell,$$

so that

$$\vec{S}(kr) = \vec{L}(kr) \sim \frac{\hbar}{2V}\frac{\sin^2(kr)}{(kr)^2}\vec{J}, \quad kr \gg 2. \quad (12)$$

Thus, the spin and OAM densities contribute equally into the total AM of a monochromatic $E1$ photon in the wave

zone. Because of the same operator structure, it is impossible to distinguish between the spin and OAM parts by any measurement in the wave zone. This result reflects well known fact that the total AM of $E1$ photon cannot be divided into the spin and OAM parts [13].

Consider now emission of a photon by the same atomic transition as above but take into consideration the natural line breadth. The time-dependent wave function of the atom-field system can be represented as follows

$$|\psi(t)\rangle = C|\psi^{(0)}\rangle + \int B(k,t)|\psi(k)\rangle dk, \tag{13}$$

where the first term corresponds to the excited atomic state and the vacuum state of the field. In turn, the second term gives the ground atomic state and a single $E1$ photon. Employing the Markov approximation then gives

$$\begin{aligned} C(t) &= e^{-i\omega_0 t-\Gamma t}, \\ B(k,t) &= -\frac{k^{3/2}}{\omega_k-\omega_0+i\Gamma}\left(1-e^{i(\omega_k-\omega_0)t-\Gamma t}\right), \end{aligned}$$

where ω_0 is the atomic transition frequency and Γ is the radiative decay width. Through the averaging of the operators (2) and (3) in the spherical wave approximation (6) over the state (13), we get

$$\langle S_z(t)\rangle = \langle L_z(t)\rangle = \frac{\hbar}{2}(1-e^{-2\Gamma t}). \tag{14}$$

Since the Markov approximation implies the long-time scale $t \geq 1/\Gamma$ [20], the last result (14) corresponds to the distances $r \geq c/\Gamma \gg c/\omega_0$ that again agree with the wave zone. This means that the spin and OAM parts contribute equally into the total AM in the wave zone independent of whether or not we take the natural line breadth into account. Effects in the near zone deserve special consideration.

Consider now quantum fluctuations of AM of $E1$ photon. Assume that the atomic transition emits a single photon in the Fock state $|1_m\rangle$, where $m = 0, \pm 1$. It is then seen that

$$\langle(\Delta J_{x,y})^2\rangle = \begin{cases} 1, & \text{if } m=0 \\ \frac{1}{2}, & \text{otherwise} \end{cases} \tag{15}$$

For all m, we have $\langle(\Delta J_z)^2\rangle = 0$. Thus, the state of $E1$ photon with projection $m = 0$ of the total AM undergoes more strong quantum fluctuations than the states with $m = \pm 1$. This means that the Fock state $|1_0\rangle$ of $E1$ photon should be less stable than $|1_\pm\rangle$.

At first sight, it may seem strange that the spin density operator of $E1$ photon in (8) has all three components. The polarization is usually associated with spin states and only two polarizations are allowed for a photon [11, 13]. In fact, there is no contradiction. The point is that the components of AM operator (9) are determined in a reference frame connected with the atomic dipole moment and that the Poynting vector not necessarily coincides with the radial direction at any point. At $r \to 0$, Poynting vector vanishes, so that $E1$ photon is created with three polarizations (two circular and one linear along the z-axis) [21]. At any $r > 0$, a local transformation of the reference frame, turning the z-axis in the direction of Poynting vector, can be determined [9]. In this local frame, there are only two polarizations. For the definition of polarization operators in the case of multipole radiation, see Ref. [22].

For the purposes of quantum information processing, the emission of photon twins by a dipole forbidden atomic transition is of high interest because the photons can be entangled in this case (e.g., see [10]). In the usual treatment, the polarization entanglement is considered. Here we examine the entanglement with respect to AM.

Assume that the excited state has the AM $j_e = 2$, while for the ground state $j_g = 0$. The two photons emitted by the transition take away the angular momentum $j = 2$. Thus, according to the classification of two-photon states [13], the radiation field can be observed in three states. Two of them are even and one is odd. Since the excited atomic state is fivefold degenerated with respect to the projection of AM m, assume that the photon twins are created by the transition

$$|j_e = 2, m_e = 0\rangle \to |j_g = 0, m_g = 0\rangle. \tag{16}$$

Then, in view of the conservation of the total projection, the even states are

$$\begin{aligned} |\psi_1\rangle &= |1_0; 1_0\rangle, \\ |\psi_2\rangle &= \frac{1}{\sqrt{2}}(|1_+; 1_-\rangle + |1_-; 1_+\rangle), \end{aligned}$$

while the odd state is

$$|\psi_3\rangle = \frac{1}{\sqrt{2}}(|1_+; 1_-\rangle - |1_-; 1_+\rangle).$$

Here $|1_m; 1_{m'}\rangle = |1_m\rangle \otimes |1_{m'}\rangle$ and the two photons move in opposite directions. The state $|\psi_3\rangle$ is the eigenstate of the Hamiltonian of atom-photon interaction

$$\begin{aligned} H &= \sum_m \omega a_m^+ a_m + \omega_0 R_{ee} \\ &+\gamma \sum_{m,m'} (R_{eg} a_m a_{m'} + a_m^+ a_{m'}^+ R_{ge}) \end{aligned} \tag{17}$$

and therefore cannot be achieved in the process of radiation. Summation in the last term in (17) is performed under the condition $m + m' = 0$ and $R_{ij} = |i\rangle\langle j|$ is the atomic operator. Hence, the eigenstate of the radiation field can be chosen in the following form

$$|\psi\rangle = \psi_1|\psi_1\rangle + \psi_2|\psi_2\rangle, \tag{18}$$

By construction, (18) represents a nonseparable two-qutrit state and may manifest entanglement [23]. Since the measure of entanglement in this case is $\mu(\psi) = |\psi_1\psi_2^2|$ [24], the state (18) is entangled if $\psi_1, \psi_2 \neq 0$. To find the state, corresponding to maximum entanglement, we

should use the variational principle of Ref. [24]. For qutrits with the $SU(3)$ dynamic symmetry of the Hilbert space, the measurements that can be performed over photons are provided by the Hermitian generators of the $SU(3)$ subalgebra in the Weyl-Heisenberg algebra (7). These generators are represented as follows

$$[M] = \begin{pmatrix} a_m^+ a_m - a_{m-1}^+ a_{m-1} \\ \frac{1}{2}(a_m^+ a_{m-1} + H.c.) \\ \frac{1}{2i}(a_m^+ a_{m-1} - H.c.) \end{pmatrix} \tag{19}$$

Here we assume cyclic permutation of the subscript m, so that $m-1=+1$ if $m=-1$. Among the three operators in the first row in (19) only two are independent.

The variational principle for maximum entanglement [24] can be expressed by the condition [25, 26]

$$\forall M \quad \langle\psi|M|\psi\rangle = 0,$$

which is valid if

$$|\psi_2| = \sqrt{2}|\psi_1| = \sqrt{2/3}. \tag{20}$$

Thus, the transition (16) gives rise to photon twins entangled with respect to the projection of AM. The two-qutrit state (18) can be maximum entangled under the condition (20). Let us stress that usually the two-qubit entangled state with respect to polarization is considered [10].

Summarizing, we have discussed the spin and OAM parts of the total AM of $E1$ photons emitted by atoms. We showed that the use of the representation of spherical waves of photons leads to a violation of commutation relations (4) for the spin and OAM. The physical result coming from Eqs. (12) and (14) is that the spin and OAM cannot be distinguished by means of any measurement in the wave zone. The use of an idealized single-mode model shows the difference in the distance dependence between spin and OAM in the near zone. We showed that the two electric dipole photons emitted by a dipole forbidden transition can manifest entanglement and even maximum entanglement with respect to the projection of AM.

The obtained results can be generalized on the case of other multipole photons. In particular, the case of magnetic dipole photons is of some considerable interest because the photon states are specified in this case by a unique OAM quantum number [13].

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