M. C. TEICH

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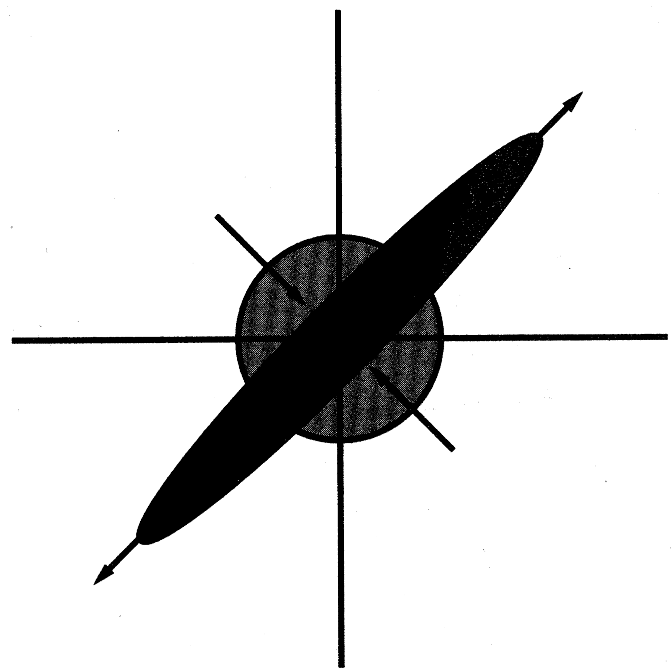

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MULTIPHOTON ABSORPTION CROSS SECTION FOR THE ENTANGLED n-PHOTON STATE

Jan Peřina, Jr.[1]

Bahaa E. A. Saleh

Malvin C. Teich

Quantum Imaging Laboratory, Department of Electrical and Computer Engineering
Boston University, 8 St. Mary's Street, Boston, MA 02215, USA

Abstract

Using a microscopic theory, we determine the entangled n-photon state generated by the nonlinear process of spontaneous n-photon parametric downconversion. An expression for the entangled n-photon absorption cross section is also obtained. The absorption cross section exhibits a linear dependence on the photon flux and depends on the entanglement times characterizing these photons. The effect of relative path delay in the beams is discussed. Effects competitive with entangled 3-photon absorption are discussed.

1 Introduction

In recent years a great deal of attention has been devoted to the study of properties of entangled two-photon states, mainly in connection with experiments based on coincidence-count measurements (violation of Bell's inequalities) [1, 2]. Effects connected with entangled three- and multiple-photon states have been discussed.

However, the nonclassical properties of such states (entanglement of photons in a state) strongly influence the behavior of photons in other cases as well. Investigations of entangled two-photon absorption [3] revealed a phenomenon called entangled two-photon transparency, which originates in the indistinguishability connected with the notion of entanglement time.

We study a generalization of entangled two-photon absorption to the case of entangled n-photon absorption. In general, entangled n-photon absorption depends on the manner in which photons in the field are entangled. In this contribution, we study the absorption of light in entangled n-photon state generated by the nonlinear process of n-photon parametric downconversion.

[1] On leave from the Joint Laboratory of Optics of Palacký University and the Academy of Sciencies of the Czech Republic, 17. listopadu 50, 772 07 Olomouc, Czech Republic.

2 Generation of entangled n-photon states

We assume that the entangled n-photon state is generated via the nonlinear process of n-photon parametric downconversion. This process can be described by the following interaction Hamiltonian [4]

$$\begin{aligned}\hat{H}_{\text{int}}(t) &= \sum_{k_1}\cdots\sum_{k_n}\chi^{(n)}(\omega_p;\omega_1,\ldots,\omega_n)\xi_p \\ &\times\frac{1}{V}\int d^3\mathbf{r}\exp(i\Delta\mathbf{k}\mathbf{r})\exp(-i\Delta\omega t)\prod_{j=1}^{n}\hat{a}^{\dagger}_{k_j}+\text{h.c.},\end{aligned} \tag{1}$$

where $\hat{a}^{\dagger}_j$ is the creation operator of the jth mode with frequency ω_j and wave vector $\mathbf{k}_j$, ξ_p is a coherent amplitude of the strong pump mode with frequency ω_p and wave vector $\mathbf{k}_p$, $\chi^{(n)}$ is the nth order susceptibility, V is the volume of the crystal, and h.c. means Hermitian conjugate. Frequency- and wave-vector mismatches are defined as $\Delta\omega=\omega_p-\sum_{j=1}^{n}\omega_j$ and $\Delta\mathbf{k}=\mathbf{k}_p-\sum_{j=1}^{n}\mathbf{k}_j$, respectively.

Because the nonlinear process is very weak, the contributions of the vacuum state and the entangled n-photon state prevail in the wavefunction describing the photon field within the crystal. That means that the use of perturbation theory in second order is justified (for details for $n=2$, see [5]). From the point of view of the statistical properties of the photon field, the process of n-photon absorption is characterized by a matrix element of the product of n positive frequency parts of the electric-field operators $\hat{E}^{(+)}_j(t_j)$ ($\hat{E}^{(+)}_j(t_j)\approx\sum_{k_j}\hat{a}_{k_j}\exp(-i\omega_j t_j)$), sandwiched between the entangled n-photon state and the vacuum state.

In our case, this element than has the form

$$\begin{aligned}\langle\text{vac}|\prod_{j=1}^{n}\hat{E}^{(+)}_j(t_j)|n\text{-photon state}\rangle &= \mathcal{N}\exp(-i\sum_{j=1}^{n}\omega_j^0 t_j) \\ &\times\frac{1}{T_{21}}\text{rect}\left(\frac{t_2-t_1}{T_{21}}\right)\prod_{j=3}^{n}\delta\left[(t_j-t_1)-\frac{T_{j1}}{T_{21}}(t_2-t_1)\right],\end{aligned} \tag{2}$$

where $\mathcal{N}$ is a normalization constant, $\text{rect}(t)$ is the rectangular function ($\text{rect}(t)=1$ for $0\le t\le 1$, $\text{rect}(t)=0$ otherwise); $\delta(t)$ is the Dirac delta function.

When deriving Eq. (2) we assumed that the central frequencies ω_j^0 and the central wave vectors $\mathbf{k}_j^0$ fulfill the phase matching conditions $\sum_{j=1}^{n}\omega_j^0=\omega_p$ and $\sum_{j=1}^{n}\mathbf{k}_j^0=\mathbf{k}_p^0$, respectively.

The entanglement times T_{ij} defined by $T_{ij}=\kappa_i-\kappa_j$ ($\kappa_i=L/v_i$, where L is the length of the crystal, v_i is the group velocity of the jth photon) and introduced in Eq. (2) characterize indistinguishability in the entangled n-photon state which is connected with the impossibility of determining the position in the crystal at which the downconverted light was created. The δ functions in Eq. (2) reflect the fact that all n photons are created at the same point in the crystal. Once we know the times at which, e.g., the two photons at ω_1 and ω_2 appear at the end of the crystal, we approximately know the position in the crystal at which all of the photons were created which determines the occurrence times of the remaining photons.

The normalization constant $\mathcal{N}$ must be chosen so that the field contains one n-photon state. In order to do that it is necessary to develop a space-time formulation of the state [6]. We further assume that the Dirac delta functions in Eq. (2) are smoothed (a finite width θ_j of the delta function whose argument contains t_j is then proportional to the inverse of the spectral width of the jth photon band). The result is

$$\mathcal{N}^2 = \frac{\hbar^n \left(\prod_{j=1}^n \omega_j^0\right) T_{21} \left(\prod_{j=3}^n \sqrt{2\pi}\theta_j\right)}{\epsilon_0^n c^{n-1} \mathcal{V} \left(\prod_{j=2}^n A_{ej}\right)}. \tag{3}$$

The symbol A_{ej} denotes the entanglement area of the photon at ω_j with respect to the photon at ω_1, which occupies a volume $\mathcal{V}$. The symbol ϵ_0 is the permittivity of vacuum, $\hbar$ is the reduced Planck constant, and c is the speed of light in vacuum.

3 Entangled n-photon absorption

The expression for the absorption cross section of light in an entangled n-photon state is derived using nth order time-dependent perturbation theory. Interaction of the photon field with the target is assumed to be described by an interaction Hamiltonian of the form

$$\hat{H}_{int} = \hat{d}\hat{E}^{(+)}(t) + \text{h.c.}, \tag{4}$$

where $\hat{d}$ is the dipole momentum operator of an electron in the target.

In order to simplify our calculations we assume the condition $T_{21} < T_{31} < \ldots < T_{n1}$, which means that the photon at ω_1 first comes at the target, the photon at ω_2 comes second, etc. The resulting expression for the absorption cross section $\sigma(T_{21}, T_{31}, \ldots, T_{n1})$, using the entangled n-photon state, becomes

$$\sigma(T_{21}, T_{31}, \ldots, T_{n1}) = 2\pi\delta\left(\varepsilon_f - \varepsilon_i - \sum_{j=1}^n \omega_j^0\right) \frac{\left(\prod_{j=1}^n \omega_j^0\right) T_{21} \left(\prod_{j=3}^n \sqrt{2\pi}\theta_j\right)}{\hbar^n \epsilon_0^n c^n \left(\prod_{j=2}^n A_{ej}\right)} \times \left| \sum_{j_{n-1},\ldots,j_1} d^n_{f j_{n-1}} \ldots d^1_{j_1 i} \frac{1 - \exp(i\Phi)}{\Phi} \right|^2, \tag{5}$$

where

$$\Phi = \sum_{l=1}^n \left(\varepsilon_{j_l} - \varepsilon_{j_{l-1}} - \omega_l^0\right) \kappa_l, \qquad (\varepsilon_0 = \varepsilon_i, \varepsilon_n = \varepsilon_f). \tag{6}$$

Here $d^j_{j_1 j_2}$ denotes the matrix element of the dipole momentum operator between the electron states j_1 and j_2 for the direction given by the jth photon polarization and ε_j denotes the frequency of the jth electron eigenstate (in particular, ε_i (ε_f) represents the frequency of the initial (final) state).

Entangled n-photon absorption is linearly dependent on the photon-flux density, which can be easily understood from the form of the square of the normalization constant $\mathcal{N}$ in Eq. (3) which is linearly dependent on the factor $c/\mathcal{V}$ which determines the photon-flux density for the "reference" mode.

We can see from Eq. (5) that the absorption cross section is a complicated function of parameters which characterize the entanglement of photons in the optical field and those connected with the structure of the target. In order to gain insight into the expression for the absorption cross section, we explicitly write it for the case of three photons as

$$\sigma(T_{21}, T_{32}) = 2\pi\delta\left(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0 - \omega_3^0\right) \frac{\omega_1^0\omega_2^0\omega_3^0\, T_{21}\sqrt{2\pi}\theta_3}{\hbar^3\epsilon_0^3 c^3 A_{e2}A_{e3}}$$

$$\times \left| \sum_{j_1,j_2} d^3_{fj_2} d^2_{j_2j_1} d^1_{j_1i} \frac{1-\exp\left(i[T_{32}(\varepsilon_f - \varepsilon_{j_2} - \omega_3^0) - T_{21}(\varepsilon_{j_1} - \varepsilon_i - \omega_1^0)]\right)}{T_{32}(\varepsilon_f - \varepsilon_{j_2} - \omega_3^0) - T_{21}(\varepsilon_{j_1} - \varepsilon_i - \omega_1^0)} \right|^2 . \tag{7}$$

Values of the absorption cross section $\sigma(T_{21}, T_{32})$ strongly depend on the entanglement times T_{21} and T_{32}. This is shown for the case of atomic hydrogen in Fig. 1. Dips occur for which the absorption cross section decreases by several orders of magnitude. This effect is called entanglement-induced transparency [3].

In the case of the entangled 3-photon state we have, in general, two entanglement times which can be adjusted to get entanglement-induced transparency. Comparing this with entanglement-induced two-photon transparency, it is interesting to suppose that, e.g., $T_{32}\Delta\omega_{ch} \approx 0$ ($\Delta\omega_{ch}$ is a characteristic detuning frequency). In this case photons at ω_2 and ω_3 have nearly the same group velocities in the crystal. The dependence of the absorption cross section $\sigma(T_{21})$ is then the same as for the entangled 2-photon state. However, in general, there are different rules for possible transitions from an initial to a final state for two- and three-photon processes.

4 Introduction of path delay

The introduction of a delay T into the path of the photon at ω_1 (the fastest one) and the assumption $T_{32}\Delta\omega_{ch} \approx 0$ leads to the following result for the absorption cross section $\sigma(T_{31}, T)$:

$$\sigma(T_{31}, T) = 2\pi\delta\left(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0 - \omega_3^0\right) \frac{\omega_1^0\omega_2^0\omega_3^0\, T_{31}\sqrt{2\pi}\theta_3}{\hbar^3\epsilon_0^3 c^3 A_{e2}A_{e3}} \mathcal{B}, \tag{8}$$

where

$$\begin{aligned} \mathcal{B} &= \left| \sum_j d^1_{fj} q^{3,2}_{ji} \frac{1-\exp[i(\varepsilon_f - \varepsilon_j - \omega_1^0)T]}{(\varepsilon_f - \varepsilon_j - \omega_1^0)T_{31}} \right. \\ &\quad \left. - \sum_j q^{3,2}_{fj} d^1_{ji} \frac{1-\exp[i(\varepsilon_j - \varepsilon_i - \omega_1^0)(T - T_{31})]}{(\varepsilon_j - \varepsilon_i - \omega_1^0)T_{31}} \right|^2 \\ &\qquad \text{for } 0 < T < T_{31}, \end{aligned} \tag{9}$$

$$\begin{aligned} &= \left| \sum_j d^1_{fj} q^{3,2}_{ji} \exp[i(\varepsilon_f - \varepsilon_j - \omega_1^0)T] \frac{1-\exp[-i(\varepsilon_f - \varepsilon_j - \omega_1^0)T_{31}]}{(\varepsilon_f - \varepsilon_j - \omega_1^0)T_{31}} \right|^2 \\ &\qquad \text{for } T > T_{31}. \end{aligned} \tag{10}$$

Quadrupole moments $q^{3,2}_{j_1j_2}$ (the notation is similar to that for dipole moments) appear as a consequence of the nearly simultaneous arrivals of photons at frequencies ω_2 and ω_3. The nearly simultaneous arrival of two entangled photons means that the interaction of this entity with an electron is characterized only by quadrupole moments, i.e., there is no competitive channel based on dipole moments. This feature creates a basis for the measurement of quadrupole moments associated with transitions between levels for which dipole moments are nonzero, which can provide a useful form of spectroscopy [6].

As indicated by the above expression for the absorption cross section $\sigma(T_{31}, T)$, there are two different regions for $\sigma(T_{31}, T)$. In particular, for $T < T_{31}$ three photons can arrive at the same time, which is impossible in the region where the time delay of the photon at ω_1 exceeds the entanglement time ($T > T_{31}$). A different behavior of $\sigma(T_{31}, T)$ in these regions is demonstrated in Fig. 2 for atomic hydrogen. The behaviour of $\sigma(T_{31}, T)$ for $T < T_{31}$ is more oscillatory as a consequence of the pairwise "interference" (see Eq. (9) for $\mathcal{B}$). The existence of two channels of the electron transition is closely related to the possibility of simultaneous arrival of all three photons. In the region with $T < T_{31}$ there are characteristic "valleys" which correspond to fixed values of the quantity $T - T_{31}$ (compare with Eq. (9) for $\mathcal{B}$).

Introduction of a nonzero path delay can lead to a resonant enhancement of the absorption (see the expression for $\sigma(T_{31}, T)$ in Eq. (9)).

The above-described features of path delay are useful when extracting information about parameters characterizing the material system [6].

5 Effects competitive with entangled 3-photon absorption

If the photon-flux density of entangled n-photon entities is large, there are competitive processes of non-entangled 3-photon absorption (each photon from a separate entity) and of doubly-entangled 3-photon absorption (two photons form one entity, the third from another entity). These processes are dependent on the third and second powers of the photon-flux density, respectively. A nonstandard linear dependence of entangled n-photon absorption on the photon-flux density thus enables us to distinguish the contributions of these various processes. The photon-flux density (I) dependence of absorption a has the form

$$a(I) = \delta_1 I + \delta_2 I^2 + \delta_3 I^3, \tag{11}$$

where δ_1 is the entangled 3-photon absorption cross section, δ_2 is the doubly-entangled 3-photon absorption cross section, and δ_3 is the non-entangled 3-photon absorption cross section.

The above absorption cross sections can be determined quantum-mechanically. But it is sufficient to restrict ourselves to a simple probabilistic model in order to determine at which photon-flux densities the processes proportional to the second and third powers of the photon flux density start to dominate.

We assume that a single photon has absorption cross section σ_s and that there is a typical relaxation time τ in the material system. We further assume that $\tau << T_e$ and $\sigma_s << A_e$, T_e being a typical entanglement time of the photons, A_e being a typical entanglement area. Strictly speaking the above assumption $\tau << T_e$ is not compatible with the above quantum model in which damping in the material system is neglected. But in spite of this, the probabilistic model is useful and provides an estimate for the values of photon-flux densities at which entangled 3-photon absorption prevails.

Simple probabilistic considerations lead to an entangled 3-photon absorption cross section δ_1 of the form

$$\delta_1 = \frac{\sigma_s^3 \tau^2}{T_e^2 A_e^2}. \tag{12}$$

The absorption cross section δ_2 for the doubly-entangled 3-photon absorption becomes,

$$\delta_2 \approx \frac{9\sigma_s^3 \tau^2}{T_e A_e}, \tag{13}$$

whereas the non-entangled 3-photon absorption is characterized by

$$\delta_3 \approx 27\sigma_s^3 \tau^2. \tag{14}$$

If we compare the contributions from these various processes, we conclude, that the process of entangled 3-photon absorption prevails at lower photon-flux densities obeying the inequality

$$I < I_{th} \approx \frac{1}{9A_e T_e}. \tag{15}$$

6 Conclusion

We have studied the properties of an entangled n-photon state generated by the process of n-photon parametric downconversion. In particular, we have found that the absorption of light in such a nonclassical state is proportional to the first power of the photon-flux density. We have determined the absorption cross section for light in an entangled n-photon state, which depends on the entanglement times of the photon field and on parameters describing the target. Entanglement-induced transparency can arise.

Introduction of an additional path delay into the path of one photon reveals the possibility of resonance enhancement of the absorption. The absorption cross section as a function of entanglement time and path delay decomposes naturally into two regions according to the possibility of the simultaneous arrival of all three photons.

A probabilistic model has been used in which the effect of nonclassical entangled 3-photon absorption dominates over other field-matter processes at lower intensities, enabling us to obtain an estimate for the threshold value of the photon-flux density.

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