

Two-photon–two-atom excitation by correlated light states

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We study the efficiency of different two-photon states of light to induce the simultaneous excitation of two atoms of different kinds when the sum of the energies of the two photons matches the sum of the energies of the two atomic transitions, while no photons are resonant with each individual transition. We find that entangled two-photon states produced by an atomic cascade are indeed capable of enhancing by a large factor the simultaneous excitation probability as compared to uncorrelated photons, as predicted some years ago by Muthukrishnan *et al.*, but that several unentangled, separable, correlated states, produced either by an atomic cascade or parametric down-conversion, or even appropriate combinations of coherent states, have comparable efficiencies. We show that the key ingredient for the increase of simultaneous excitation probability is the presence of strong frequency anticorrelation and neither time correlation nor time-frequency entanglement.

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I. INTRODUCTION

Quantum entanglement and its inherent nonlocal properties are among the most fascinating and challenging features of the quantum world. In addition, entanglement plays a central role in quantum information [1–5]. Since its first description in the 1930s [6] and in spite of the decisive contribution of Bell [7] and the subsequent experimental studies [8], entanglement appears as a rather mysterious and puzzling property, even for bipartite systems. In particular, distinguishing between effects related to genuine entanglement and those related to the quantum correlations measured on a single quantum observable is a difficult task [9], as can be seen, for example, by the great number of papers about quantum discord [10,11]. Some time ago, a paper was published [12] which showed that some entangled states are able to induce transitions in quantum systems that factorized states cannot excite. The physical problem studied in that paper is therefore a good test bench to examine in detail in a simple situation the role of entanglement and of correlations not related to entanglement. This is the purpose of the present paper.

The problem under consideration is the probability of two-photon–two-atom (2P2A) excitation in the situation where the two atoms are of different species and have different transition frequencies, and the light to which the atoms are submitted is in general non-resonant for each one but resonant for the system of two atoms. Two-photon absorption by single atoms or molecules has been studied since 1931 [13] and remains a current subject of theoretical and experimental research [14–18]. When the atoms have more than one intermediate state, many important features, including

cross-section cancellation and enhancement, are observed [15]. These features have recently been shown to be applicable in characterizing the two-photon quantum states [18].

It has also been known for a long time that two-photon resonant excitation of two different atoms is indeed possible when the two atoms are interacting [19]. A nearly monochromatic light beam will have a resonant two-photon absorption peak when tuned across the average frequency of the two atoms. Different experiments have since confirmed this theoretical prediction [20–22]. In addition to a direct potential interaction between the atoms, such as the dipole-dipole, cooperative 2P2A has also been predicted for pairs of atoms inside an optical cavity [23]. In this case the physical interaction is mediated by the radiation background surrounding the atoms. Reference [12] addresses the case of two-photon absorption in absence of interaction between the two atoms, with the excitation made using some particular entangled state of light. The conclusion of the authors of [12] is that *in some situations entanglement can replace a real physical interaction*, which is a far-reaching statement and an important physical property related to entanglement. Surprisingly, this question did not attract much attention for several years. The same subject was also considered, but in the context of spin entanglement in electron paramagnetic resonance, by Salikhov [24]. More recently [25,26], the related problem of interaction with pairs of broadband spectrum photons has been discussed.

In this paper, we consider in more detail the problem of two-atom excitation in order to determine the exact role of entanglement in the process. More precisely, we determine the probability of 2P2A excitation by different multimodal

states of light. From these results we draw conclusions on the respective role of entanglement and of correlations not related to entanglement in such a process. Section II gives the general framework in which the problem is treated. Section III derives from a second-order perturbation theory the relevant transition probability. Results for various different two-photon states, introduced in Sec. IV, are given in Secs. V and VI. Finally, in Sec. VII we discuss different hypotheses for the physical origin of the enhancement of the 2P2A process.

II. THE MODEL

Let us first describe the model we are using and the notations. We consider two different two-level atoms labeled (1) and (2), having ground and excited states $|g_i\rangle$ and $|e_i\rangle$ ($i = 1, 2$), corresponding Bohr frequencies ω_i , and spontaneous emission rates γ_i , interacting with a quantized field. We assume that the mean atomic excitation time is much shorter than the lifetimes of the two excited atoms so that we can consider that the two excited states have infinite lifetimes ($\gamma_{1,2} \simeq 0$) and keep forever their excitation. For the sake of simplicity we will assume that the light source is far from the atoms, so that the only nonempty modes are plane-wave modes having a single propagation direction Oz and a single polarization. Thus one can use annihilation operators depending only on the frequency $a(\omega_\ell) = a_\ell$. Under the rotating-wave approximation the Hamiltonian of the system is then given by

$$\begin{aligned} H &= H_{\text{atom}}^0 + H_f^0 + V, \\ H_{\text{atom}}^0 &= \hbar\omega_1 b_1^\dagger b_1 + \hbar\omega_2 b_2^\dagger b_2, \quad b_i = |g_i\rangle\langle e_i|, \\ H_f^0 &= \sum_\ell \hbar\omega_\ell a_\ell^\dagger a_\ell, \quad [a_\ell, a_{\ell'}^\dagger] = \delta_{\ell, \ell'}, \\ V &= \hbar b_1^\dagger \sum_\ell f_1(\omega_\ell) a_\ell + \hbar b_2^\dagger \sum_\ell f_2(\omega_\ell) a_\ell + \text{H.c.}, \end{aligned} \quad (1)$$

where $f_i(\omega_\ell) = -id_i \sqrt{\omega_\ell/2\hbar\epsilon_0 SL} e^{i\omega_\ell z_i/c} = f_{i\ell} e^{i\omega_\ell z_i/c}$, $f_{i\ell}$ being a slowly varying function of the photon frequency. d_i is the electric dipole matrix element of atom i , S the transverse section of the beam which is focused on the atoms, z_i the position of atom i , and L the length of the quantization box, the mode density in terms of frequencies ω_ℓ being $2\pi c/L$. For simplicity, we will set $z_1 \simeq z_2 \simeq 0$ and hence will not consider propagation effects. In this case, we may approximate $f_i(\omega_\ell) \simeq f_i(\omega_i)$.

The evolution of the whole system is described by a unitary operator U , and the input light ρ_0 can be either a pure two-photon state $|\Psi_\mu\rangle = \sum_{kq} c_{kq}^\mu |1_k, 1_q\rangle$ or a mixed state in its spectral decomposition form $\rho_0 = \sum_\mu p_\mu |\Psi_\mu\rangle\langle\Psi_\mu|$. The probability of 2P2A excitation is given in this case by

$$\begin{aligned} P(t) &= \sum_\mu p_\mu \langle\Psi_\mu, g_1, g_2|U^\dagger(t)|0, e_1, e_2\rangle \\ &\quad \times \langle 0, e_1, e_2|U(t)|\Psi_\mu, g_1, g_2\rangle. \end{aligned} \quad (2)$$

The exact expression of the evolution operator unfortunately is not easy to obtain. To simplify our discussion, we will use lowest-order perturbation theory, which is a good approximation for two-photon states.

III. SECOND-ORDER PERTURBATION THEORY

Due to the weak coupling between the light field and the two atoms, the leading term in the evolution is $U^{(2)} = -\hbar^{-2} e^{-it(H_{\text{atom}}^0 + H_f^0)/\hbar} \int_0^t d\tau \int_0^\tau ds \tilde{V}(\tau) \tilde{V}(s)$, where \tilde{V} is the coupling term in the interaction picture. One has

$$\langle e_1 e_2 | U^{(2)} | g_1 g_2 \rangle = e^{-it(\omega_1 + \omega_2 + H_f^0/\hbar)} \sum_{mn} a_m a_n \mathcal{A}_{mn}, \quad (3)$$

$$\mathcal{A}_{mn} = f_1(\omega_m) f_2(\omega_n) \frac{1 - e^{i(\omega_1 - \omega_m)t}}{\omega_m - \omega_1} \frac{1 - e^{i(\omega_2 - \omega_n)t}}{\omega_n - \omega_2}. \quad (4)$$

Therefore the leading term of the coexcitation probability (48) reads

$$P(t) \simeq \sum_{jkmn} \mathcal{A}_{jk}^* \mathcal{A}_{mn} \text{Tr}(a_j^\dagger a_k^\dagger a_m a_n \rho_0). \quad (5)$$

In the case of a continuous frequency distribution of photons, one must replace the sum by an integral:

$$\begin{aligned} \langle e_1 e_2 | U^{(2)} | g_1 g_2 \rangle &= e^{-it(\omega_1 + \omega_2 + H_f^0/\hbar)} \frac{L^2}{4\pi^2 c^2} \\ &\quad \times \iint d\omega_m d\omega_n a(\omega_m) a(\omega_n) \mathcal{A}_{mn}, \end{aligned} \quad (6)$$

where $a(\omega)$ is the annihilation operator of a monochromatic photon of frequency ω .

Note that the coefficient \mathcal{A}_{mn} is the product of two factors which represent the response of each atom to the field. When time t goes to infinity, these two factors behave roughly like two independent Dirac δ functions centered on the atom resonances (we will detail this argument in Sec. VI). The 2P2A excitation probability is indeed induced by the individual wings of the incident light spectrum, which are resonant with the atoms. Consequently, if one photon is absorbed by one atom, there is no reason why the second photon should be absorbed by the second atom in a way correlated to the absorption of the first photon. In other words, the 2P2A excitation phenomenon has no reason *a priori* to have a resonant behavior when the 2P2A resonance condition $\omega_1 + \omega_2 = \omega_m + \omega_n$ is fulfilled.

However, as the process is nonlinear and involves two atoms, it can be enhanced by taking advantage of correlation effects between the atoms or between the photons:

(1) A first possibility consists in introducing an interaction between the two atoms. Let $\omega_u \simeq \omega_1 + \omega_2$ be the maximal Bohr frequency of the two-atom system. If one photon with frequency ω_k is absorbed, then the two atoms will be more likely to absorb another photon with frequency $\omega_u - \omega_k$ in a resonant two-photon process [19,27]. This was experimentally demonstrated in [20–22] using nearly degenerate photon pairs.

(2) A second possibility is to use correlated photons to interact with the two atoms. Let us consider a source that emits correlated photons. If a photon is absorbed by one atom, then the remaining atom will interact with its correlated photon with a higher probability, leading to enhanced 2P2A resonance (we detail this argument and the kind of correlation needed in Sec. VII).

We will now discuss the ideas by having a closer look at different possible light states likely to induce such a 2P2A transition.

IV. ENTANGLED, CORRELATED-SEPARABLE, AND FACTORIZED TWO-PHOTON STATES

Before we go further, let us discuss the different kinds of two-photon states that we will consider in the following. Starting from entangled pure quantum state $|\Psi\rangle$, having a density matrix $\rho_0 = |\Psi\rangle\langle\Psi|$ of matrix elements $\rho_{kk'qq'} = \langle 1_k, 1_q | \rho_0 | 1_{k'}, 1_{q'} \rangle$, one can construct others that have the same mean energy and the same single-photon spectrum, and hence that would give the same transition probabilities for a single-photon resonance. We choose two special cases that will allow a quantitative evaluation of the role of correlations:

The first one is defined as

$$\rho_1 = \sum_{k,q} \rho_{kkqq} |1_k, 1_q\rangle\langle 1_k, 1_q|. \quad (7)$$

It is the diagonal part of ρ_0 . It has lost any temporal field coherence and is time independent. It is actually a *correlated-separable state* [28], which results from the “disentanglement” of the previous one. It gives rise, however, to correlations between its two parties.

The second one is defined as

$$\rho_2 = \sum_{k,q} \sum_{q'} \rho_{kkq'q'} |1_k\rangle\langle 1_k| \otimes \sum_{k'} \rho_{k'k'qq} |1_q\rangle\langle 1_q|. \quad (8)$$

This is a fully factorized state, which does not give rise to any correlation whatsoever. These states will induce 2P2A excitation with respective probabilities $P_1(t)$ and $P_2(t)$.

The two diagonal density matrices (ρ_1, ρ_2) describe two cw fields while the entangled pure state ρ_0 describes a pulse. As a result, at a time t , the flow of energy having interacted with atoms in each state is different. However, as shown in Appendix B, this quantity is quite the same in each state when $t = L/c$. For comparison, we will take $t = L/c$ through the whole paper.

We are of course interested in cases where an increase in the excitation probability is expected. When this happens with $P(L/c) \gg P_2(L/c)$ and $P(L/c) \gg P_1(L/c)$, then entanglement is indeed the key to efficient 2P2A transition, whereas if $P(L/c) \simeq P_1(L/c) \gg P_2(L/c)$, correlations, of quantum or classical origin, are more important than entanglement in the present problem.

V. TWO-PHOTON-TWO-ATOM EXCITATION INDUCED BY DIFFERENT TWO-PHOTON QUANTUM FIELD STATES

We will now examine the efficiency of various multimode light states for the simultaneous excitation of the two atoms.

A. Two quasimonochromatic uncorrelated photons

Let us begin by the simplest case: two uncorrelated photon wave packets of mean frequencies ω_α (much closer to ω_1) and ω_β , and respective spectral widths γ_α and γ_β much bigger than the detecting atom spectral widths γ_1 and γ_2 , emitted by two uncorrelated atoms excited at the same time in the past and arriving at the detecting atom’s position at $t = 0$, described

therefore by the two-photon state $|\psi^{11}\rangle$ with

$$|\psi^{11}\rangle = \sum_k \frac{g_\alpha(\omega_k)}{\omega_k - \omega_\alpha + i\gamma_\alpha} |1_k\rangle \otimes \sum_q \frac{g_\beta(\omega_q)}{\omega_q - \omega_\beta + i\gamma_\beta} |1_q\rangle. \quad (9)$$

It is the tensor product of two single-photon wave packets [29] of duration γ_α^{-1} and γ_β^{-1} . In the calculation of the probability amplitude in (5), we will replace the sum over modes by the double integral $(L^2/4\pi^2c^2) \iint_0^\infty d\omega_k d\omega_q$, extend each integration domain to the whole real axis, and use the residue theorem [30]. When $\gamma_\alpha t \gg 1$ and $\gamma_\beta t \gg 1$ while keeping $\gamma_1 t \ll 1$ and $\gamma_2 t \ll 1$, the transition probability amplitude is

$$A^{11} \simeq \frac{L^2 f_1(\omega_1) f_2(\omega_2) g_\alpha(\omega_1) g_\beta(\omega_2)}{c^2(\omega_1 - \omega_\alpha + i\gamma_\alpha)(\omega_2 - \omega_\beta + i\gamma_\beta)}. \quad (10)$$

The coefficients $g_\mu(\omega_\ell)$ can be taken as constant, and the normalization of the two-photon state imposes

$$g_\alpha g_\beta = \frac{2c\sqrt{\gamma_\alpha\gamma_\beta}}{L}, \quad (11)$$

so that the transition probability P^{11} is

$$P^{11} = \frac{P_0 \gamma_\alpha \gamma_\beta}{[(\omega_1 - \omega_\alpha)^2 + \gamma_\alpha^2][(\omega_2 - \omega_\beta)^2 + \gamma_\beta^2]}, \quad (12)$$

where $P_0 = d_1^2 d_2^2 \omega_1 \omega_2 / \hbar^2 \varepsilon_0^2 c^2 S^2 = 36\pi^2 \gamma_1 \gamma_2 c^4 / \omega_1^2 \omega_2^2 S^2$, in which we have used the expression for the spontaneous emission rate $\gamma_i = d_i^2 \omega_i^3 / (6\pi \epsilon_0 \hbar c^3)$ [29,30].

1. Double resonance

According to (12), the best transition probability will be achieved when the photons are separately resonant with the two atoms $\omega_\alpha = \omega_1$ and $\omega_\beta = \omega_2$, namely, the double-resonance (DR) condition, under which the transition probability is then equal to

$$P_{DR}^{11} = \frac{P_0}{\gamma_\alpha \gamma_\beta}, \quad (13)$$

which can be written in a more general way

$$P_{DR}^{11} = \frac{P_0}{S_{fr}}, \quad (14)$$

where S_{fr} is the effective area of frequency distribution $|c_{kq}|^2$ in the (ω_k, ω_q) plane (see Fig. 2). This result turns out to be general and implies that all pure states having the same effective areas S_{fr} , entangled or not, will produce the same doubly resonant transition probability. Thus we regard (13) as a universal result under the double-resonance condition, and its value will serve as a reference for all subsequent transition probabilities.

2. Two-photon-two-atom resonance

When none of the two photons are resonant with the two atoms but where the sum of their two energies almost matches the sum of the two atomic energies $\omega_\alpha + \omega_\beta \simeq \omega_1 + \omega_2$, the transition probability (12) has in this case no resonant variation as a function of the 2P2A detuning:

$$\delta = \omega_\alpha + \omega_\beta - \omega_1 - \omega_2. \quad (15)$$

When $\delta = 0$ the transition probability is

$$P_{2P2A}^{11} = \frac{P_0 \gamma_\alpha \gamma_\beta}{\Delta^4} = P_{DR}^{11} \frac{\gamma_\alpha^2 \gamma_\beta^2}{\Delta^4}, \quad (16)$$

where Δ is the smallest frequency mismatch between the emitting atom frequencies, and the detecting atom frequencies are supposed to be much larger than the atomic widths. Without loss of generality, we have taken $\Delta = |\omega_\alpha - \omega_1| = |\omega_2 - \omega_\beta|$.

We then conclude that the special case of 2P2A excitation probability by uncorrelated photons is also nonzero for any couple of frequencies $\omega_\alpha, \omega_\beta$; thus such a two-photon transition turns out not to be disallowed but simply induced by the wings of the two single-photon frequency resonances. It is therefore very weak, as witnessed by the Δ^{-4} variation of probability.

B. Two photons produced by an atomic cascade

Let us now envision the case considered in [12] of a two-photon light state produced by a three-level atom excited at a given time in the upper state that cascades down to the ground state on two successive transitions of Bohr frequencies successively equal to ω_α and ω_β . The corresponding spontaneous emission rates are γ_α and γ_β . We assume that the emitted light is wholly directed in the Oz direction of atoms (1) and (2) (by means of a parabolic mirror, for example) and can only arrive after $t = 0$. It is described by a two-photon wave packet with a coefficient c_{kq}^{cas} equal, at a time t long compared to the lifetimes of the two transitions, to [12,30]

$$c_{kq}^{cas} = \frac{g_\alpha(\omega_k) g_\beta(\omega_q)}{[\omega_k + \omega_q - \omega_\alpha - \omega_\beta + i\gamma_\alpha][\omega_q - \omega_\beta + i\gamma_\beta]}. \quad (17)$$

Here this entangled nonstationary state is produced by a cascade so that the photon of frequency ω_q always arrives just after the photon of frequency ω_k . In addition, the probability to have photons of frequency sum $\omega_k + \omega_q$ close to $\omega_\alpha + \omega_\beta$ is high. We have therefore an entangled state which is not only correlated in time but also anticorrelated in frequency. It is the time-energy analog of the position-momentum entangled state introduced by Einstein-Podolsky-Rosen, or of the field quadrature entangled state [3,30,31].

Using the residue theorem to integrate over the frequencies ω_k and ω_q , the transition probability amplitude reads exactly as follows:

$$A^{cas} = \frac{L^2}{c^2} \frac{g_\alpha(\omega_1) g_\beta(\omega_2) f_1(\omega_1) f_2(\omega_2)}{\omega_{\beta 2} - \delta - i(\gamma_\beta - \gamma_\alpha)} \times \left[\frac{1 - e^{-(\gamma_\beta + i\omega_{\beta 2})t}}{\omega_{\beta 2} - i\gamma_\beta} - \frac{1 - e^{-(\gamma_\alpha + i\delta)t}}{\delta - i\gamma_\alpha} \right] + (1 \leftrightarrow 2). \quad (18)$$

When $\gamma_{1,2}^{-1} \gg t \gg \gamma_{\alpha,\beta}^{-1}$, the four decaying terms in Eq. (18) are negligible, leading to a compact expression

$$A^{cas} = -\frac{L^2}{c^2} \frac{f_1(\omega_1) f_2(\omega_2)}{\delta - i\gamma_\alpha} \times \left[\frac{g_\alpha(\omega_1) g_\beta(\omega_2)}{\omega_{\beta 2} - i\gamma_\beta} + \frac{g_\alpha(\omega_2) g_\beta(\omega_1)}{\omega_{\beta 1} - i\gamma_\beta} \right], \quad (19)$$

where $\omega_{\mu\nu} = \omega_\mu - \omega_\nu$ is the frequency difference between frequency ω_μ and frequency ω_ν ; $\mu, \nu = \alpha, \beta, 1, 2, k, q$.

1. Double resonance

Starting from (19) and keeping only the largest term, one obtains in this case for the probability amplitude,

$$A_{DR}^{cas} \simeq \frac{L^2 f_1(\omega_1) f_2(\omega_2) g_\alpha(\omega_1) g_\beta(\omega_2)}{c^2 \gamma_\alpha \gamma_\beta}. \quad (20)$$

Using the same assumption as in the previous calculation, one finds for the probability

$$P_{DR}^{cas} = \frac{P_0}{\gamma_\alpha \gamma_\beta} = P_{DR}^{11}. \quad (21)$$

It is time independent because we are considering times much longer than the two-photon pulse of duration $\gamma_\alpha^{-1} + \gamma_\beta^{-1}$. As it is equal to the probability obtained with uncorrelated photons, we conclude that entanglement does not help in the fully resonant case, but does not harm either.

2. Two-photon-two-atom resonance

Let us now turn to the 2P2A resonance case. One obtains in this case for the probability,

$$P_{2P2A}^{cas} \simeq \frac{L^2}{4c^2} \frac{P_0}{\delta^2 + \gamma_\alpha^2} \left[\frac{g_\alpha(\omega_1) g_\beta(\omega_2)}{\omega_2 - \omega_\beta} + \frac{g_\alpha(\omega_2) g_\beta(\omega_1)}{\omega_1 - \omega_\beta} \right]^2. \quad (22)$$

This expression, already obtained in [30], shows that for this state the probability has indeed a resonant character around the 2P2A resonance $\delta = 0$. The transition probability P_{2P2A}^{cas} at the exact 2P2A resonance is then

$$P_{2P2A}^{cas} \simeq \frac{P_0}{\gamma_\alpha \gamma_\beta} \frac{\gamma_\beta^2}{\Delta^2} = P_{DR}^{11} \frac{\gamma_\beta^2}{\Delta^2}. \quad (23)$$

One therefore finds that the transition probability is in the present case smaller than P_{DR}^{11} by a factor $(\gamma_\beta/\Delta)^2$ at exact 2P2A resonance, as expected because one is now less resonant than in the double-resonance case. One finds more importantly that P_{2P2A}^{cas} is larger than P_{2P2A}^{11} , i.e., than in the two-uncorrelated-photon case, by a factor $(\Delta/\gamma_\alpha)^2$, which can be very large. This enhancement of the 2P2A transition probability is the main result of [12]. Entanglement may indeed significantly enhance the two-photon-two-atom process. To the best of our knowledge no experiment has been undertaken to show such a striking effect.

It must be emphasized that the present considerations do not imply that the atom cascade entangled state is the only one likely to produce such a significant increase in the transition probability. This is the reason why we will now consider other light quantum states which may also be of interest in the present problem.

C. Correlated and factorized states analogous to the atomic cascade

Let us now consider the two states that have the same energy and the same spectrum that we have introduced in Sec. IV, namely, the correlated-separable state,

$$\rho_1 = \left(\frac{2c}{L} \right)^2 \sum_{kq} \frac{\gamma_\beta}{(\omega_{q\beta}^2 + \gamma_\beta^2)} \frac{\gamma_\alpha}{[(\omega_{q\beta} + \omega_{k\alpha})^2 + \gamma_\alpha^2]} \times |1_k, 1_q\rangle \langle 1_k, 1_q|, \quad (24)$$

and the factorized state,

$$\rho_2 = \left(\frac{2c}{L}\right)^2 \left(\sum_k \frac{\gamma_\alpha + \gamma_\beta}{\omega_{k\alpha}^2 + (\gamma_\alpha + \gamma_\beta)^2} |1_k\rangle\langle 1_k| \right) \otimes \left(\sum_q \frac{\gamma_\beta}{\omega_{q\beta}^2 + \gamma_\beta^2} |1_q\rangle\langle 1_q| \right). \quad (25)$$

The first one corresponds to an atomic cascade for which the starting time is random, thereby averaging to zero all the off-diagonal time-dependent terms in the density matrix; the second one characterizes a mixed state with two uncorrelated photons having the same spectrum as the initial cascade state. They give rise to the following transition probabilities:

$$P_1 \simeq P_0 \frac{\gamma_\alpha \gamma_\beta}{\delta^2 + \gamma_\alpha^2} \left(\frac{1}{(\omega_1 - \omega_\beta)^2} + \frac{1}{(\omega_2 - \omega_\beta)^2} \right) \frac{t^2}{(L/c)^2}, \quad (26)$$

$$P_2 \simeq P_0 \gamma_\beta (\gamma_\alpha + \gamma_\beta) \left(\frac{1}{(\omega_1 - \omega_\beta)^4} + \frac{1}{(\omega_2 - \omega_\beta)^4} \right) \frac{t^2}{(L/c)^2}. \quad (27)$$

At exact 2P2A resonance, we have $P_1 \simeq P_{DR}^{11} \gamma_\beta^2 c^2 t^2 / (\Delta^2 L^2)$ and $P_2 \simeq P_{DR}^{11} \gamma_\alpha \gamma_\beta^2 (\gamma_\alpha + \gamma_\beta) c^2 t^2 / (\Delta^4 L^2)$. At any time t , one finds $P_1 \gg P_2$, since the spectral widths are much smaller than single-photon detunings. This fact shows that correlations indeed play an important role in the efficiency of the excitation.

Note that P_1 and P_2 depend on time, as can be expected in a situation where the detecting atoms, which have an infinite lifetime, are submitted to a stationary quantum state, and therefore to cw light. In order to compare P_1 and P_2 to P_{2P2A}^{cas} [Eq. (22)], which is induced by a pulse of light, we need to fix an interaction time t . It is shown in Appendix B that the two atoms are submitted to the same energy flow at time $t = L/c$. One then obtains at this time and at exact resonance

$$P_1 \simeq P_{DR}^{11} \frac{\gamma_\beta^2}{\Delta^2} \simeq P_{2P2A}^{cas}. \quad (28)$$

We thus find the result that *a correlated-separable state like ρ_1 can induce the 2P2A transition as efficiently as the entangled cascade state*. This statement constitutes the main result of the present paper.

Let us stress that ρ_1 , though not entangled, indeed has genuine quantum properties, being a mixture of single-photon states which are highly nonclassical. It exhibits strong correlations that we will study in more detail in Sec. VI.

D. Two-photon state produced by parametric down-conversion

The two-photon state $|\Psi_{pdc}\rangle$ produced by nondegenerate spontaneous parametric down-conversion (SPDC), which has been under wide and in-depth investigation for many years, is another important photon source in quantum optics. Because of its $\chi^{(2)}$ nonlinearity, a nonlinear crystal submitted to a pulsed pump field of central frequency $\omega_\alpha + \omega_\beta$ and narrow bandwidth σ_α emits a signal field (central frequency ω_α) and an idler field (central frequency ω_β). Let σ_β be the frequency width of the phase-matching curve. For the sake of computational simplicity we will use a Gaussian approximation for both the laser line shape and the phase-matching curve. The crystal generates in such a case an entangled state which is described

by a wave packet with a coefficient c_{kq}^{pdc} [32] given by

$$c_{kq}^{pdc} = \mathcal{N} e^{-\frac{(\omega_{k\alpha} + \omega_{q\beta})^2}{2\sigma_\alpha^2} + i(\omega_{k\alpha} + \omega_{q\beta})t_0} \left(e^{-\frac{\omega_{k\alpha}^2 + \omega_{q\beta}^2}{2\sigma_\beta^2}} + i e^{-\frac{\omega_{k\beta}^2 + \omega_{q\alpha}^2}{2\sigma_\beta^2}} \right), \quad (29)$$

where \mathcal{N} is the normalized coefficient, satisfying

$$\left(\frac{L}{2\pi c}\right)^2 \mathcal{N}^2 \frac{2\pi\sigma_\alpha\sigma_\beta^2}{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}} = 1.$$

In expression (29), we have assumed that the pump laser pulse had a Gaussian temporal shape centered at time $t_0 \gg \sigma_\alpha^{-1} + \sigma_\beta^{-1}$ to provide most of the photons a chance to interact with the two detecting atoms. The factor i in the second term originates from a relative phase (depending on the birefringence), which is set to be $\pi/2$ for the sake of simplicity in our case.

Here we will also extend the double integral to the whole plane and find, when t is sufficiently large [33], the transition probability

$$P^{pdc} = \pi P_0 \frac{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}}{\sigma_\alpha\sigma_\beta^2} e^{-\frac{\delta^2}{\sigma_\alpha^2}} \left(e^{-\frac{\omega_{1\alpha}^2 + \omega_{2\beta}^2}{2\sigma_\beta^2}} + e^{-\frac{\omega_{2\alpha}^2 + \omega_{1\beta}^2}{2\sigma_\beta^2}} \right)^2. \quad (30)$$

The two mixed two-photon states ($\rho_1^{pdc}, \rho_2^{pdc}$), defined in Sec. IV and pertaining to the pure SPDC two-photon state (29), are

$$\rho_1^{pdc} = \mathcal{N}^2 \sum_{kq} e^{-\frac{(\omega_{k\alpha} + \omega_{q\beta})^2}{\sigma_\alpha^2}} \left(e^{-\frac{\omega_{k\alpha}^2 + \omega_{q\beta}^2}{\sigma_\beta^2}} + e^{-\frac{\omega_{k\beta}^2 + \omega_{q\alpha}^2}{\sigma_\beta^2}} \right) \times |1_k, 1_q\rangle\langle 1_k, 1_q|, \quad (31)$$

$$\rho_2^{pdc} = \pi \frac{c^2}{L^2} \frac{\zeta}{\sigma_\beta^2} \left[\sum_k \left(e^{-\zeta \frac{\omega_{k\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{k\beta}^2}{\sigma_\beta^2}} \right) |1_k\rangle\langle 1_k| \right] \otimes \left[\sum_q \left(e^{-\zeta \frac{\omega_{q\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{q\beta}^2}{\sigma_\beta^2}} \right) |1_q\rangle\langle 1_q| \right], \quad (32)$$

where $\zeta = 1 + \sigma_\beta^2/(\sigma_\alpha^2 + \sigma_\beta^2)$. The first one corresponds to an SPDC process in which all the off-diagonal time-dependent terms in the density matrix are averaged to zero by random processes, while the second one characterizes a mixed state with two uncorrelated photons having the same spectrum than the initial SPDC state. When t is sufficiently large, their corresponding transition probabilities read

$$P_1^{pdc} = \pi P_0 \frac{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}}{\sigma_\alpha\sigma_\beta^2} e^{-\frac{\delta^2}{\sigma_\alpha^2}} \left(e^{-\frac{\omega_{1\alpha}^2 + \omega_{2\beta}^2}{\sigma_\beta^2}} + e^{-\frac{\omega_{1\beta}^2 + \omega_{2\alpha}^2}{\sigma_\beta^2}} \right) \times \left(\frac{t}{L/c}\right)^2, \quad (33)$$

$$P_2^{pdc} = \pi \frac{P_0}{2} \frac{\zeta}{\sigma_\beta^2} \left(e^{-\zeta \frac{\omega_{1\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{1\beta}^2}{\sigma_\beta^2}} \right) \left(e^{-\zeta \frac{\omega_{2\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{2\beta}^2}{\sigma_\beta^2}} \right) \times \left(\frac{t}{L/c}\right)^2. \quad (34)$$

We will once again take $t = L/c$ to be able to compare in a fair way the pulsed and cw excitations through the rest of the following discussions.

1. Double resonance

Taking $\omega_\alpha = \omega_1, \omega_\beta = \omega_2$ and keeping the largest term, one finds the probability

$$P_{DR}^{pdc} = P_{1,DR}^{pdc} \simeq \pi P_0 \frac{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}}{\sigma_\alpha \sigma_\beta}, \quad (35)$$

which once again implies that entanglement is not active in enhancing the transition probability in the double-resonance case.

One also finds $P_{DR}^{pdc} \simeq P_{DR}^{11}$ when $\sigma_\alpha = \gamma_\alpha, \sigma_\beta = \gamma_\beta$. In the following we will take this correspondences of spectral widths for comparison. Henceforth, P_{DR}^{pdc} or P_{DR}^{11} will be regarded as a reference in the discussions related to SPDC two-photon states.

2. Two-photon–two-atom resonance

The transition probability P^{pdc} has indeed a resonant character around $\delta = 0$ according to (30). At the exact 2P2A resonance, its value is assessed at

$$P_{2P2A}^{pdc} \simeq P_{DR}^{11} e^{-2\Delta^2/\sigma_\beta^2}, \quad (36)$$

which is much smaller than for the atom cascade state because the factor Δ^2/σ_β^2 enters now as an exponent in a Gaussian function and the detuning Δ is much greater than the spectral widths.

For the factorized, uncorrelated state ρ_2^{pdc} , the transition probability in this case reads

$$\begin{aligned} P_{2,2P2A}^{pdc} &\simeq P_{DR}^{11} (1 + 2\sigma_\beta^2/\sigma_\alpha^2)^{-1/2} e^{-2\zeta\Delta^2/\sigma_\beta^2} \\ &\lesssim e^{-2\Delta^2/(\sigma_\alpha^2 + \sigma_\beta^2)} P_{2P2A}^{pdc}. \end{aligned} \quad (37)$$

Thus P_{2P2A}^{pdc} is much greater than the probability given by the factorized state because of the scale factor $e^{2\Delta^2/(\sigma_\alpha^2 + \sigma_\beta^2)}$. So we obtain in the parametric down-conversion configuration the same conclusion as the one drawn in [12] for the atomic cascade: the entangled state $|\Psi_{pdc}\rangle$ is much more efficient for inducing a 2P2A resonance than the factorized, uncorrelated state.

For the correlated-separable state ρ_1^{pdc} , the transition probability reads

$$P_{1,2P2A}^{pdc} \simeq P_{DR}^{11} e^{-2\Delta^2/\sigma_\beta^2} \simeq P_{2P2A}^{pdc}. \quad (38)$$

Thus one has $P_{1,2P2A}^{pdc} \gg P_{2,2P2A}^{pdc}$. The same conclusion is found as in the cascade case: the correlated-separable state is as efficient as the entangled state to boost the 2P2A resonance. The fact that $P_{1,2P2A}^{pdc}$ is much larger than $P_{2,2P2A}^{pdc}$, and $P_{1,2P2A}^{pdc} \simeq P_{2P2A}^{pdc}$, once again shows that correlations, which are not necessarily related to entanglement, indeed play a crucial role in the efficiency of the excitation.

VI. ENHANCEMENT OF 2P2A RESONANCE FOR MORE GENERAL CLASSES OF LIGHT STATES

We have so far studied interesting but specific states of light and showed an enhancement effect for some of them, entangled or correlated-separable. It would be interesting to consider now more general classes of light states.

A. Light pulses starting at a given time

Let us go back to the initial equations (4) and (5). They contain functions such as $[1 - \exp(i\omega_{1m}t)]/\omega_{1m}$. When $t \rightarrow \infty$, as explained in Appendix A, even though this function does not act as a Dirac function when it is applied to integrations with any function, it indeed tends to $2i\pi\delta(\omega_1 - \omega_m)$ if applied to functions of ω_m that have a Fourier transform, which is strictly zero for $t < 0$. Such will be the case here.

The initial two-photon light state $|\Psi\rangle$ is the pure state,

$$|\Psi\rangle = \sum_{kq} c_{kq} |1_k, 1_q\rangle, \quad (39)$$

that describes a “switched-on” light which is not vacuum only after time $t = 0$. One can then use the δ function approximation. The probability that the two atoms are found in the excited state at long times compared to the pulse duration is now

$$P \simeq \frac{P_0 L^2}{4 c^2} |c_{12} + c_{21}|^2. \quad (40)$$

Mathematically, if $|c_{12}| \sim |c_{21}|$, this interference, which has been studied in the literature [16], may lead to strong variations according to the relative phase. According to the Cauchy-Schwartz inequality, one has

$$0 \leq P \leq 2 \left(\frac{P_0 L^2}{4 c^2} (|c_{12}|^2 + |c_{21}|^2) \right). \quad (41)$$

However, physically speaking, only one component between c_{12} and c_{21} dominates in the expression (40). This is because we have assumed that the quantities $\omega_1, \omega_2, \omega_\alpha, \omega_\beta$ are sufficiently separated from each other but with a small 2P2A detuning $\delta \simeq 0$; as a result, ω_1 should be closer to one of the central frequencies of the fields than to the other ones. Under this condition, one has

$$P \simeq \frac{P_0 L^2}{4 c^2} (|c_{12}|^2 + |c_{21}|^2). \quad (42)$$

The correlated and factorized states ρ_1, ρ_2 analogous to the initial state $|\Psi\rangle\langle\Psi|$ give rise to the following 2P2A transition probabilities:

$$P_1 = \frac{P_0}{4} t^2 (|c_{12}|^2 + |c_{21}|^2), \quad (43)$$

$$P_2 = \frac{P_0}{4} t^2 \sum_{mn} (|c_{1n} c_{m2}|^2 + |c_{2n} c_{m1}|^2). \quad (44)$$

The enhancement of the 2P2A transition probability is characterized by the quotient G_p between P and P_1 at $t = L/c$:

$$G_p = \frac{P}{P_1} \Big|_{t=L/c} = \frac{|c_{12} + c_{21}|^2}{|c_{12}|^2 + |c_{21}|^2}. \quad (45)$$

Thus one finds $0 \leq G_p \leq 2$. The maximum value of 2 is achieved when $c_{12} = c_{21}$.

One has $G_p \simeq 1$ under the physical conditions we stated before. That is, the entangled and the correlated-separable state yield almost equal transition probabilities. This implies that the conclusion that we had drawn in the special previous cases is valid for a large class of two-photon states: correlated states are as efficient as entangled states in 2P2A coexcitation when they have delivered the same amount of energy to the two atoms.

Another important discriminability index is the ratio between the two transition rates P_1 and P_2 :

$$G_{12} = \frac{|c_{12}|^2 + |c_{21}|^2}{\sum_{mn} (|c_{1n}c_{m2}|^2 + |c_{2n}c_{m1}|^2)}. \quad (46)$$

The value of the enhancement factor G_{12} can be used as a witness for the correlation needed in such a problem.

Note in addition that, while P is sensitive to possible destructive interference effects between c_{12} and c_{21} , P_1 is not. Therefore the enhancement effect as indicated by G , and due to correlations not related to entanglement, turns out to be more “robust” than the one related to it.

B. Coherent states

So far we have only considered two-photon states of different shapes, which are all strongly nonclassical objects, as they are produced by spontaneous emission or parametric fluorescence, which are specifically quantum processes with no classical equivalent. But one can also envision superpositions of two-mode coherent states of the form

$$|\Psi_{coh}\rangle = \sum_{kq} c_{kq} |\alpha(\omega_k)\rangle \otimes |\alpha(\omega_q)\rangle, \quad (47)$$

where $|\alpha(\omega_k)\rangle$ is the coherent state $|\alpha\rangle$ in the mode of frequency ω_k , α being the same complex number for all modes.

As the field state is no longer a two-photon state, the calculation of the transition probability must be redone from the beginning. One finds that the probability of 2P2A excitation is given by

$$P_{coh}(t) = \langle \Psi_{coh} | \langle g_1, g_2 | U^\dagger(t) | e_1, e_2 \rangle \langle e_1, e_2 | U(t) | g_1, g_2 \rangle | \Psi_{coh} \rangle. \quad (48)$$

By using the approximation $\langle \alpha | 0 \rangle \approx 0$ valid for $|\alpha| \gg 1$, one finally finds

$$P_{coh}(t) = |\alpha|^4 P(t), \quad (49)$$

where $P(t)$ is the probability (5) obtained for two-photon states. Apart from the energy scaling factor $|\alpha|^4$, the conclusions of the previous paragraphs hold in the present case, which looks much more classical than the previously studied ones, as such states can be produced by classical means.

VII. WHAT KIND OF CORRELATION IS REQUIRED TO ENHANCE THE 2P2A TRANSITION PROBABILITY?

We have found in the previous sections that the 2P2A transition probability depends crucially on the specific state of light used for the excitation, even when all the considered states have the same energy spectrum. The question we address now is the physical origin of an enhanced transition probability. We have seen that entangled and unentangled states may

give comparable results, so a first answer to the question is obviously that entanglement is not at the origin of the effect but rather is some kind of correlation effect which is shared by entangled and unentangled states.

Candidates likely to play a role in the present problem are time correlation and frequency correlation. We will now examine them successively.

A. Temporal correlation effect

It is well characterized by the cross-second-order correlation function $g_{\times}^{(2)}(t, \tau)$:

$$g_{\times}^{(2)}(t, \tau) = \frac{\text{Tr}[\rho_0 \hat{E}_{\alpha}^{(-)}(\tau) \hat{E}_{\beta}^{(-)}(t) \hat{E}_{\beta}^{(+)}(t) \hat{E}_{\alpha}^{(+)}(\tau)]}{\text{Tr}[\rho_0 \hat{E}_{\alpha}^{(-)}(t) \hat{E}_{\alpha}^{(+)}(t)] \text{Tr}[\rho_0 \hat{E}_{\beta}^{(-)}(t) \hat{E}_{\beta}^{(+)}(t)]}. \quad (50)$$

Assuming that the amplitude of the single-photon electric field is a smooth function of ω_k , one gets the following for the pure state $|\Psi\rangle = \sum_{kq} c_{k,q} |1_k, 1_q\rangle$:

$$g_{\times}^{(2)}(t, \tau) = \left| \sum_{kq} c_{k,q} e^{-i\omega_k \tau - i\omega_q t} \right|^2. \quad (51)$$

It is the modulus square of two-time Fourier transform of the two-photon state.

(1) In the case of the cascade state (17),

$$g_{\times}^{(2)}(t, \tau) = \left(\frac{L}{2\pi c} \right)^2 \frac{\gamma_{\alpha} \gamma_{\beta}}{\pi^2} \theta(\tau) \theta(t - \tau) e^{-2\gamma_{\alpha} \tau - 2\gamma_{\beta} (t - \tau)}, \quad (52)$$

$\theta(t)$ being the step function. We notice here a time asymmetry between t and τ , expected in the case of a cascade in which the ω_{α} photon is always emitted before the ω_{β} photon.

(2) For the SPDC state (29),

$$g_{\times}^{(2)}(t, \tau) = \frac{2}{\mathcal{N}^2} [1 + \sin \omega_{\alpha\beta} (t - \tau)] \times \exp \left[-\frac{\sigma_{\beta}^2 (t - \tau)^2}{2} - \frac{2\sigma_{\alpha}^2 \sigma_{\beta}^2}{\sigma_{\alpha}^2 + 2\sigma_{\beta}^2} \left(t_0 - \frac{t + \tau}{2} \right)^2 \right]. \quad (53)$$

As can be seen in Fig. 1, $g_{\times}^{(2)}(t, \tau)$ is in both cases significant only very close to the diagonal, which implies that both states exhibit strong temporal correlations, as expected. The width of the diagonal, which gives the characteristic time of this correlation, is equal to $\gamma_{\beta}^{-1} (\sigma_{\beta}^{-1})$ in both the cascade and SPDC cases.

It is easy to see that for the correlated-separable states (24) and (31), there is no time dependence for $g_{\times}^{(2)}(t, \tau)$, and hence no temporal correlation, as expected from a cw time-averaged state in which the photons arrive at any time. It is also the case for the coherent states (47). As these states give 2P2A transition probabilities comparable to the entangled state, we must conclude that the temporal correlation is not the physical origin of the enhancement effect, nor is the time ordering of the photons present in the cascade state. The physical reason is that, as we have neglected their spontaneous emission, the two detecting atoms have an infinite memory time, and hence they can be excited separately at any time.

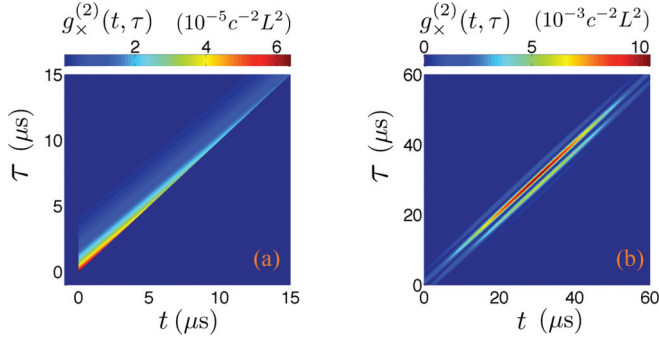


FIG. 1. (Color online) Plots of the cross-temporal correlation function $g_x^{(2)}(t, \tau)$. (a) The atom cascade two-photon state, with $\gamma_\alpha = 0.05$ MHz and $\gamma_\beta = 0.5$ MHz; (b) the SPDC two-photon state, in which the pulse takes place around $t_0 = 30 \mu\text{s}$, and with $\sigma_\alpha = 0.05$ MHz, $\sigma_\beta = 0.5$ MHz, $\omega_{\beta\alpha} = 2$ MHz. Note the (t, τ) asymmetry in the first figure and fringes in the second one are due to interferences from two temporal processes. In both plots, one finds significant temporal correlations along the diagonal line. In a real condition, the value of $\omega_{\beta\alpha}$ should be much greater, leading to a poorer graphic representation for interference patterns.

B. Frequency correlation effect

It is well characterized by the cross-second-order frequency correlation function $g_x^{(2)}(\omega, \omega')$,

$$g_x^{(2)}(\omega, \omega') = \frac{\text{Tr}[\rho_0 \hat{E}_\alpha^{(-)}(\omega') \hat{E}_\beta^{(-)}(\omega) \hat{E}_\beta^{(+)}(\omega) \hat{E}_\alpha^{(+)}(\omega')]}{\text{Tr}[\rho_0 \hat{E}_\alpha^{(-)}(\omega') \hat{E}_\alpha^{(+)}(\omega')] \text{Tr}[\rho_0 \hat{E}_\beta^{(-)}(\omega) \hat{E}_\beta^{(+)}(\omega)]}, \quad (54)$$

equal in the pure-state case to $|c(\omega, \omega')|^2$ and to $\rho(\omega, \omega')$ in the mixed-state case.

This quantity is plotted in Fig. 2 for the cascade and SPDC states, either entangled, correlated-separable, or factorized. One observes that the frequency correlation functions take significant values only on the antidiagonal for the left-side plots, which implies that the corresponding states exhibit strong frequency anticorrelations. This is not the case for the right-side plots. The width of the antidiagonal, which gives the characteristic width of the frequency anticorrelation, is equal to $\gamma_\alpha(\sigma_\alpha)$ in both the cascade and SPDC entangled and correlated-separable cases.

Let us note that the entangled cascade and SPDC states are the only ones in our list exhibiting simultaneously time correlations and frequency anticorrelations: one has in these states EPR-like correlations, revealed by a violation of the time-energy Heisenberg inequality [31,34] when $\gamma_\alpha/\gamma_\beta$ (or $\sigma_\alpha/\sigma_\beta$) $\ll 1$.

The important point to notice is that such a frequency anticorrelation exists for all the states which exhibit 2P2A resonance enhancement, and is not present for the states which do not give rise to this effect. We are therefore led to the conclusion that *the property needed to enhance the 2P2A excitation is precisely the presence of strong frequency anticorrelations in the quantum state.*

This conclusion, that we have demonstrated for the two specific examples considered in the first sections of this paper, is far more general, as can be seen in the expression of the probability written for any switched-on two-photon state.

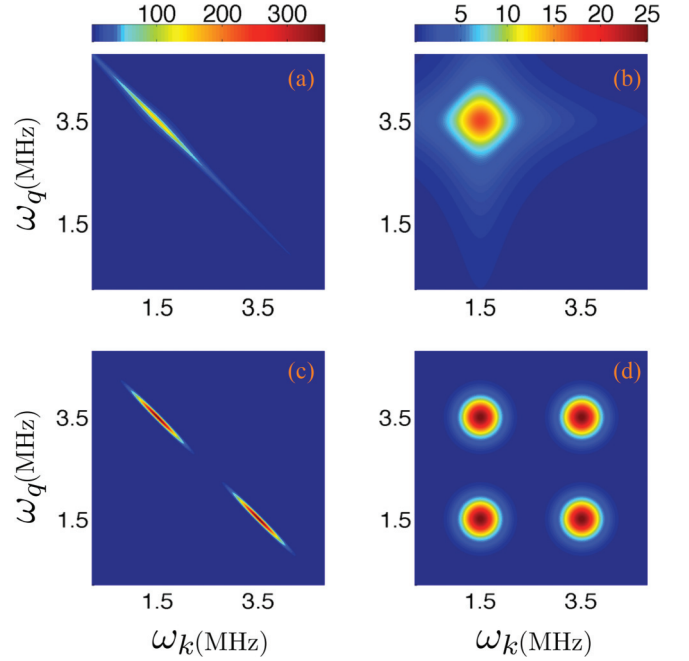


FIG. 2. (Color online) Plots of the cross-frequency correlation function $g_x^{(2)}(\omega_k, \omega_q)$: (a) entangled, correlated-separable, and coherent cascade states; (b) factorized cascade state; (c) entangled, correlated-separable, and coherent SPDC states; and (d) factorized SPDC state. In all plots $\gamma_\alpha = \sigma_\alpha = 0.05$ MHz, $\gamma_\beta = \sigma_\beta = 0.5$ MHz, $\omega_\alpha = 1.5$ MHz, and $\omega_\beta = 3.5$ MHz. The color codes, in the unit of c^2/L^2 , on the top left (right) are shared by (a) and (c) (b and d). The left-side plots exhibit strong frequency anticorrelations along the line $\omega_k + \omega_q = \omega_\alpha + \omega_\beta$, while in the right-side plots, one finds no such a correlation. The SPDC two-photon source is nondegenerate and each photon has two distribution peaks; thus one sees two bright spots in the left side bottom plot and four bright spots in the factorized case in the right-side bottom plot. In a real condition, the distances of the peaks in the bottom plots are much greater, and the sizes of spots are much smaller.

Equations (37), (38), and (39) indeed show that the probability of 2P2A excitation is proportional to the component of the density matrix of the two-photon state corresponding to the existence of one photon with frequency ω_1 and one photon with frequency ω_2 . This gives a simple interpretation of the problem: there is 2P2A excitation only when each photon of the two-photon state is resonant with the atomic transition of the atom it excites. This is expected, since we are considering that the atomic excited states have a very long lifetime, and therefore very narrow linewidths. Since the spectrum of each photon of the source has a much larger bandwidth, the probability of excitation is small. If the photons are not correlated in frequency, the probability of double excitation is proportional to the product of the probabilities that each photon has the corresponding transition frequency, and this yields a very small transition probability. But when the photons are anticorrelated in frequency such that the sum of their frequencies is equal to the sum of the transition frequencies of the atoms, when one photon is resonant with one atomic transition, the correlated photon will be automatically resonant with the other transition, and the probability of 2P2A transition will in general be much higher than in the noncorrelated case.

We can say that the 2P2A transition occurs with a higher probability when the sum of the photon frequencies is found inside a small interval around the sum of the atomic transition frequencies, so that the enhancement is associated with the inverse of the variance of the $|c_{kq}|^2$ distribution in the direction of the diagonal.

VIII. CONCLUSION

We can now answer the question raised in the Introduction about the role of entanglement in the two-photon excitation process considered in this paper. We have shown that what is necessary for the enhancement of the transition probability is neither quantum entanglement nor temporal correlations, but rather frequency anticorrelation, which can be due to the presence of entanglement in the state, but also to correlations that are not related to entanglement. This result brings new light to the problem of assessing the exact role of quantum correlations and quantum entanglement in physical processes.

As for any nonlinear process, such as two-photon absorption in a single atom [35], 2P2A transition probability can be modified by changing the quantum state of light, and therefore the enhancement effect that we have studied in this paper is due to the partial optimization of the quantum state. The absorption of two photons by the two atoms occurs when each photon is resonant with the atomic transition of the atom that absorbs it. This has a greater probability to happen in frequency-anticorrelated photon states than in uncorrelated states.

We have not treated in this paper the important question of characterizing in a quantitative way the frequency correlation relevant to the present enhancement and relating it to its classical or quantum character through various quantum correlation witnesses such as the quantum discord. It will be addressed in a subsequent paper, together with the important question of the full optimization of the quantum state with respect to the 2P2A probability maximization, given a constant spectral energy distribution.

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APPENDIX A : IS $[\exp(-i\omega t) - 1]/(2i\pi\omega)$ A GOOD APPROXIMATION OF THE δ FUNCTION?

Let us note $s_t(\omega)$ the function $[\exp(-i\omega t) - 1]/(2i\pi\omega)$. One can also write it as $s_t(\omega) = -\sin \omega t/(2\pi\omega) + i(1 - \cos \omega t)/(2\pi\omega)$. Whereas the real part of $s_t(\omega)$ is a sinc function which tends indeed to a δ function when $t \rightarrow \infty$, the imaginary part, not being a peaked function whose area is constant, is not an approximation of the δ function. So, in general, $s_t(\omega)$ does not tend to the δ distribution when it acts on the general set

of integrable functions; however, it can be so on a smaller set of functions. This set includes, for example, all the odd functions in ω , a subset which is not relevant for the present paper. We show in this appendix that $s_t(\omega)$ behaves also as a δ function when it acts on functions which have a Fourier transform, which is strictly zero before $t = 0$.

Let us consider a function $F(t)$ that is zero for $t < 0$ and admits a well-behaved Fourier transform $f(\omega)$. Then

$$f(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt F(t) e^{i\omega t} = \frac{1}{2\pi} \int_0^{\infty} dt F(t) e^{i\omega t}, \quad (\text{A1})$$

$$F(t) = \int_{-\infty}^{\infty} d\omega f(\omega) e^{-i\omega t}, \quad (\text{A2})$$

where $f(\omega)$ is absolutely integrable, which excludes functions such as $1/(\omega + i\gamma)$ from the present discussion. Let us now calculate the integral:

$$\begin{aligned} I &= \int_{-\infty}^{\infty} d\omega \frac{\exp(-i\omega t) - 1}{\omega} f(\omega) \\ &= i \int_0^t d\tau \int_{-\infty}^{\infty} d\omega f(\omega) \exp(-i\omega\tau) \\ &= i \int_0^t d\tau \theta(\tau) F(\tau). \end{aligned} \quad (\text{A3})$$

Then $I \rightarrow i \int_0^{\infty} d\tau F(\tau) = 2\pi i f(0)$ when $t \rightarrow \infty$. This proves that $s_t(\omega)$ acts as a δ function for the set of functions that have a Fourier transform strictly null for $t < 0$.

APPENDIX B : WHY DO WE TAKE $t = L/c$ IN THE COMPARISON OF TRANSITION PROBABILITIES?

In order to compare the probabilities of transitions induced by pulsed and cw light in a fair way, we must be careful to take the same amount of energy flow $\mathcal{F}(t)$ on the detecting atoms in both cases. This quantity is nothing else than the integral over time and transverse section S of the Poynting vector. It is equal to, at a given time t and for a state ρ ,

$$\begin{aligned} \mathcal{F}(t) &= 2\varepsilon_0 c S \int_0^t \text{Tr}[\rho \hat{E}^{\dagger}(\tau) \hat{E}(\tau)] d\tau \\ &\simeq \hbar\omega \frac{c}{L} \int_0^t \text{Tr}[\rho \hat{b}^{\dagger}(\tau) \hat{b}(\tau)] d\tau, \end{aligned} \quad (\text{B1})$$

where $\hat{b}(\tau) = \sum_m \hat{a}_m \exp(-i\omega_m \tau)$ and ω is the mean frequency of the state under consideration.

For any diagonal density matrix (DDM), since $\text{Tr}[\rho_{DDM} \hat{b}^{\dagger}(\tau) \hat{b}(\tau)] = 2$ is time independent, one finds a linear relationship between the energy flow and time t ,

$$\mathcal{F}_{DDM}(t) = 2\hbar\omega \frac{ct}{L}. \quad (\text{B2})$$

For any entangled pure state $|\Psi\rangle = \sum_{kq} c_{kq} |1_k, 1_q\rangle$,

$$\begin{aligned} \text{Tr}[|\Psi\rangle\langle\Psi| \hat{b}^{\dagger}(\tau) \hat{b}(\tau)] &= \sum_k \left| \sum_q c_{kq} e^{-i\omega_q \tau} \right|^2 \\ &+ \sum_q \left| \sum_k c_{kq} e^{-i\omega_k \tau} \right|^2. \end{aligned} \quad (\text{B3})$$

The energy flow at time t is

$$\mathcal{F}_\Psi(t) = \int_0^t d\tau \text{Tr}[\rho_\Psi \langle \Psi | \hat{b}^\dagger(\tau) \hat{b}(\tau) | \Psi \rangle] \approx \int_{-\infty}^t d\tau \text{Tr}[\rho_\Psi \langle \Psi | \hat{b}^\dagger(\tau) \hat{b}(\tau) | \Psi \rangle], \quad (\text{B4})$$

when most photons arrive at the detecting atoms after $t = 0$. One assumes that at sufficiently large time t (much greater than the temporal coherence length of the field), the photons in state $|\Psi\rangle$ have fully interacted with the detecting atoms; therefore one extends t to $+\infty$ without introducing notable error. By using the Parseval identity, one has

$$\begin{aligned} \mathcal{F}_\Psi(t) &\approx \hbar\omega \frac{c}{L} \int_{-\infty}^{\infty} d\tau \text{Tr}[\rho_\Psi \langle \Psi | \hat{b}^\dagger(\tau) \hat{b}(\tau) | \Psi \rangle] = \hbar\omega \frac{c}{L} \int_{-\infty}^{\infty} d\tau \left[\sum_k \left| \sum_q c_{k,q} e^{-i\omega_q \tau} \right|^2 + \sum_q \left| \sum_k c_{k,q} e^{-i\omega_k \tau} \right|^2 \right] \\ &= \hbar\omega \left[\sum_{kq} |c_{kq}|^2 + \sum_{kq} |c_{kq}|^2 \right] = 2\hbar\omega, \end{aligned} \quad (\text{B5})$$

as expected. By comparison with Eq. (B2), one finds that at time $t = L/c$, the energies supplied by the cw field and by the pulse are equal. Under this situation, one can make legitimate comparisons between the corresponding transition probabilities.

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