

Inducing Disallowed Two-Atom Transitions with Temporally Entangled Photons

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Two uncoupled two-level atoms cannot be jointly excited by classical light under general circumstances, due to destructive interference of excitation pathways in two-photon absorption. However, with temporally entangled light, two-atom excitation is shown possible. Photons arising from three-level cascade decay are intrinsically ordered in time of emission. This field correlation induces a joint resonance in the two-atom excitation probability via suppression of one of the time-ordered excitation pathways. The relative gain in two-photon absorption increases with the time-frequency entanglement.

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Entangled states of light have novel spectroscopic and interferometric properties which are based on multiphoton correlation measurements [1]. In the case of two photons, such measurements are characterized by the intensity correlation function [2]

$$G^{(2)} = \langle \hat{E}^-(\mathbf{r}_i, t_1) \hat{E}^-(\mathbf{r}_j, t_2) \hat{E}^+(\mathbf{r}_j, t_2) \hat{E}^+(\mathbf{r}_i, t_1) \rangle, \quad (1)$$

which gives the joint probability of absorbing two photons at positions \mathbf{r}_i and \mathbf{r}_j , and times t_1 and t_2 , respectively. A microscopic model for a detection system that makes a two-photon measurement is the joint excitation of two independent, two-level atoms. In this Letter, we take a closer look at this basic problem from the point of view of understanding the effects of source field entanglement on two-photon absorption. In particular, we are interested in the effects of time-frequency entanglement on the two-atom excitation probability under conditions of two-photon resonance.

Nearly 20 years ago, it was demonstrated [3] that the entangled states produced by an optical parametric amplifier (OPA) exhibit, for weak fields, a *linear* intensity dependence in the coincident absorption probability, rather than the usual quadratic dependence expected for a two-photon process. Theoretical treatments of the problem [3–6] explained the effect as due to the correlated nature of the two-photon state: the absorption of the signal photon within some coherence window automatically implies the absorption of the idler photon, as these “travel” together. However, this point of view does not readily distinguish between quantum entanglement, as found in a pure two-photon state, and classical correlation, as exists with temporally copropagating pulses.

In this Letter, we report a *qualitative* distinction between quantum and classical light that arises from the time asymmetry that is intrinsic to the two-photon state vector produced by successive decay of a three-level cascade system [7–9], one of Nature’s fundamental sources of entangled photons. A similar asymmetry can

be introduced into an OPA source by splitting the signal and idler photons produced by a type-II down converter using a polarizing beam splitter and delaying one of the photons with respect to the other before recombining them at an ordinary beam splitter. The time asymmetry in the entangled state is the key ingredient that enables the joint excitation of two noninteracting atoms, an event that is not possible to accomplish with classical light under general circumstances. We believe that the results of our Letter can be realized by utilizing two ions in a trap, as was done previously to demonstrate Young’s interference fringes for single-photon scattering [10]. The proposed two-photon experiment can be done with either a cascade or an OPA source.

To understand the effect, consider first a classical source of bichromatic light, with center frequencies ω_α and ω_β , incident on two atoms with transition frequencies ω_1 and ω_2 . If the fields are nonoverlapping in time, it is not surprising that two-photon resonance plays no role and the absorption rate goes to zero in the limit of large one-photon detunings. However, and this is a key point, even when the fields overlap, we find that two-photon

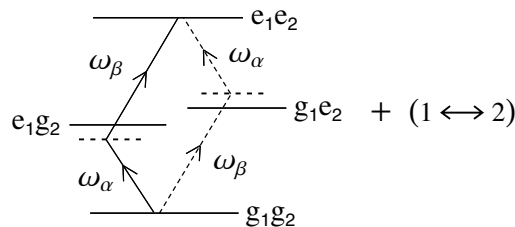


FIG. 1. Two two-level atoms (ground state g_i and excited state e_i) are driven by bichromatic light along two pathways, depending on which frequency ω_α or ω_β is absorbed first (denoted by the solid or dashed arrows), and for two pairings, depending on which atom absorbs which frequency (exchanged in $1 \leftrightarrow 2$). For each pairing, the time-ordered pathways interfere destructively to cancel classical two-photon absorption, assuming $\omega_\alpha - \omega_i \neq \omega_\beta - \omega_j$.

absorption vanishes in general. This comes about from destructive interference of time-ordered excitation pathways corresponding to which photon is absorbed first (see Fig. 1). Consider the second-order Fermi golden rule for the two-atom excitation probability:

$$P_{e_1 e_2}^{(2)} \propto \left| \frac{1}{\omega_\alpha - \omega_1} + \frac{1}{\omega_\beta - \omega_2} + (1 \leftrightarrow 2) \right|^2 \delta(\Delta_{\alpha\beta,12}), \quad (2)$$

where the δ function enforces energy conservation for times long compared to the inverse of the two-photon detuning: $\Delta_{\alpha\beta,12} = \omega_\alpha + \omega_\beta - \omega_1 - \omega_2$. We see that when $\Delta_{\alpha\beta,12} = 0$, and provided the one-photon detunings $\omega_\alpha - \omega_i$ and $\omega_\beta - \omega_j$ are distinct and nonzero, the sum of each pair of terms inside the square vanishes, and hence $P_{e_1 e_2}^{(2)} \rightarrow 0$. That is, as long as there are distinct excitation pathways corresponding to the order in which the photons are absorbed (α first or β first), they interfere destructively to cancel two-photon absorption.

The absence of two-photon absorption in the two-atom system is intuitive from the point of view that the atoms are not coupled and there is no physical mechanism to enforce a joint resonance in the excitation spectrum. However, if the atoms are coupled in some way, for example, through a dipole-dipole interaction [11,12] or through a common interaction with a quantized cavity field mode [13], a two-photon resonance at the sum frequency $\omega_\alpha + \omega_\beta$ appears in the absorption spectrum. This can be observed as photon bunching in the two-atom fluorescence [14]. Alternately, as two dipole-interacting atoms approach each other, the equivalent Dicke three-level model acquires a cooperative shift of the intermediate level b that enables two-photon absorption [15].

The question we pose in this Letter is whether, even when the atoms are *not* interacting—and are far apart—we can see a two-photon resonance in the excitation spectrum owing to the initial entanglement between the photons at the source. We note that this is the first time we are seeing a fundamental process such as two-photon absorption produced by time-entangled photons in non-interacting systems. The nonclassicality of this effect can be related to an Einstein-Podolsky-Rosen–type criterion for time-frequency entanglement, namely, that the sum frequency of the two photons cannot be defined to a precision better than, of order, the inverse of the time separation between the emissions. For the cascade state, this points to the ratio of linewidths of upper and intermediate levels as the measure of the degree of entanglement, and therefore, as we will show, a quantitative measure of the reported effect.

We consider time-frequency-entangled photons produced in cascade decay of an atomic three-level system [1,7–9], as shown in Fig. 2. If the source is located at \mathbf{r}_0 , this results in the two-photon state vector

$$|\alpha, \beta\rangle = \sum_{p,q} \varphi_{pq} |1_p, 1_q\rangle; \quad \varphi_{pq} = \frac{h_{p\alpha} h_{q\beta} \exp[-i(\mathbf{p} + \mathbf{q}) \cdot \mathbf{r}_0]}{(\omega_p + \omega_q - \omega_\alpha - \omega_\beta + i\gamma_\alpha)(\omega_q - \omega_\beta + i\gamma_\beta)}, \quad (3)$$

where p and q represent the mode frequencies, wave vectors, and polarizations of the emitted photons (the latter contained in $h_{p\alpha}$ and $h_{q\beta}$), ω_α and ω_β are the center frequencies, and γ_α and γ_β are the half-widths of the upper two levels in the decaying atom. Since φ_{pq} cannot be written as a product of the functions of p and q , the two photons are said to be nonseparable. In the limit $\gamma_\beta \gg \gamma_\alpha$, the photons are maximally entangled in frequency; i.e., the sum frequency is well defined but the individual frequencies are not.

To describe the entanglement in the time domain, we calculate the intensity correlation function in Eq. (1). For a general two-photon state $|\mu, \nu\rangle$, this can be written as

$$G^{(2)} \propto |\psi_{\mu\nu}^{(2)}(\mathbf{r}_i, \mathbf{r}_j; t_1, t_2) + \psi_{\nu\mu}^{(2)}(\mathbf{r}_i, \mathbf{r}_j; t_1, t_2)|^2, \quad (4)$$

where $\psi_{\mu\nu}^{(2)}(\mathbf{r}_i, \mathbf{r}_j; t_1, t_2)$ is the two-photon correlation amplitude corresponding to absorption of photons μ and ν , respectively, at positions \mathbf{r}_i and \mathbf{r}_j , at times t_1 and t_2 :

$$\psi_{\mu\nu}^{(2)}(\mathbf{r}_i, \mathbf{r}_j; t_1, t_2) \equiv \sum_{p,q} g_{pi} g_{qj} \exp[i(\mathbf{p} \cdot \mathbf{r}_i + \mathbf{q} \cdot \mathbf{r}_j)] \times \exp[-i(\omega_p t_1 + \omega_q t_2)] \varphi_{pq}, \quad (5)$$

where the mode index p (q) is associated with photon μ (ν), and g_{ki} is a constant that describes the coupling strength between mode k and detector i . Using φ_{pq} from Eq. (3) for the cascade state, we find [1]

$$\psi_{\alpha\beta}^{(2)} = K \theta(\tau_\alpha) \exp[-(i\omega_\alpha + i\omega_\beta + \gamma_\alpha)\tau_\alpha] \times \theta(\tau_\beta - \tau_\alpha) \exp[-(i\omega_\beta + \gamma_\beta)(\tau_\beta - \tau_\alpha)], \quad (6)$$

where $K = \kappa_{\alpha i} \kappa_{\beta j} / \Delta r_i \Delta r_j$, $\kappa_{\alpha i}$ and $\kappa_{\beta j}$ are coupling

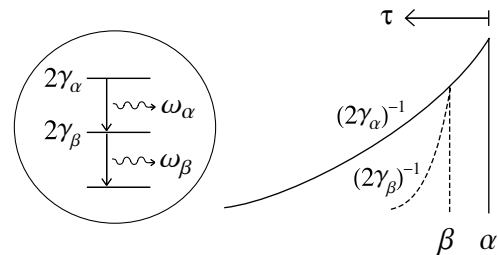


FIG. 2. The temporal profiles of two photons emitted by a cascade source illustrate time-frequency entanglement: the solid curve represents the marginal probability $P(\tau_\alpha)$; the dashed curve represents the conditional probability $P(\tau_\beta|\tau_\alpha)$. See Eqs. (7) and (8). The intrinsic time ordering of the photons, α first, followed by β , suppresses the dashed excitation pathway in Fig. 1, inducing joint two-atom excitation.

constants, $\Delta r_i = |\mathbf{r}_i - \mathbf{r}_0|$ is the distance from the source to the detector i , and $\tau_\alpha = t_1 - \Delta r_i/c$ and $\tau_\beta = t_2 - \Delta r_j/c$ are the retarded times for α and β for this term. Note that the function $\theta(\tau_\beta - \tau_\alpha)$ dictates a strict ordering of the retarded times for the two-photon absorption; hence for a given set of coordinates $(\mathbf{r}_i, \mathbf{r}_j; t_1, t_2)$, one of the two terms in Eq. (4) goes to zero. To see the temporal entanglement, consider the marginal probability $P(\tau_\alpha)$, obtained by integrating over τ_β the quantity $P(\tau_\alpha, \tau_\beta) = |\psi_{\alpha\beta}^{(2)}|^2$, and an associated conditional probability, $P(\tau_\beta|\tau_\alpha) = P(\tau_\alpha, \tau_\beta)/P(\tau_\alpha)$. We find that

$$P(\tau_\alpha) = 2\gamma_\alpha \theta(\tau_\alpha) \exp(-2\gamma_\alpha \tau_\alpha), \quad (7)$$

$$P(\tau_\beta|\tau_\alpha) = 2\gamma_\beta \theta(\tau_\beta - \tau_\alpha) \exp[-2\gamma_\beta(\tau_\beta - \tau_\alpha)]. \quad (8)$$

That is, the absorption of α turns on at $\tau_\alpha = 0$ and decays slowly at the rate γ_α , while the absorption of β turns on at $\tau_\beta = \tau_\alpha$ and decays rapidly at the rate γ_β (see time profiles in Fig. 2). Thus, the two photons arrive in strict succession, α followed by β , with the time interval between the absorptions going to zero when $\gamma_\beta \gg \gamma_\alpha$, i.e., the limit of large frequency entanglement.

With this background in mind, consider the two-atom excitation problem where the atoms are located at fixed positions \mathbf{r}_1 and \mathbf{r}_2 . In the dipole and rotating-wave approximations, the atom-field coupling in the interaction picture is given by the Hamiltonian

$$\begin{aligned} \hat{V}_I(t) = & -\sum_k g_{k1} \exp(i\mathbf{k} \cdot \mathbf{r}_1) \hat{\sigma}_1^\dagger(t) \hat{a}_k(t) + \text{H.c.} \\ & -\sum_k g_{k2} \exp(i\mathbf{k} \cdot \mathbf{r}_2) \hat{\sigma}_2^\dagger(t) \hat{a}_k(t) + \text{H.c.}, \end{aligned} \quad (9)$$

where $\hat{\sigma}_i^\dagger(\hat{\sigma}_i)$ and $\hat{a}_k^\dagger(\hat{a}_k)$ are atomic and field raising (lowering) operators that have the usual exponential time dependence in the interaction picture, namely, $\hat{\sigma}_i(t) = \hat{\sigma}_i \exp(-i\omega_i t)$ and $\hat{a}_k(t) = \hat{a}_k \exp(-i\omega_k t)$. We assume that both atoms start off in the ground state, $|g_1 g_2\rangle$, and that the field is in the two-photon state $|\alpha, \beta\rangle$. From second-order perturbation theory, the state of the system after two interaction events (at times $t_1 < t_2$) is given by

$$|\Psi^{(2)}\rangle = \int_0^t dt_2 \int_0^{t_2} dt_1 \hat{V}_I(t_2) \hat{V}_I(t_1) |g_1, g_2\rangle |\alpha, \beta\rangle. \quad (10)$$

The two-atom excitation probability $P(\omega_1, \omega_2) = |A(\omega_1, \omega_2)|^2$ corresponds to both atoms being in the excited state, $|e_1, e_2\rangle$, and the field in the vacuum state, $|0\rangle$. Thus, to lowest order, the joint excitation amplitude $A(\omega_1, \omega_2)$ is given by the projection

$$A(\omega_1, \omega_2) = \{\langle e_1, e_2 | \langle 0 | \Psi^{(2)} \rangle\}. \quad (11)$$

As an aside, consider first the ‘‘classical’’ case where the two photons are frequency separable and monochromatic, i.e., $|\alpha, \beta\rangle \rightarrow |1_\alpha, 1_\beta\rangle$. Then, only two operators \hat{a}_α and \hat{a}_β contribute to each mode sum in Eq. (9). Using the

abbreviations $|i\rangle = |g_1, g_2\rangle |1_\alpha, 1_\beta\rangle$ and $|f\rangle = |e_1, e_2\rangle |0\rangle$, we have, for this case,

$$\begin{aligned} \langle f | \hat{V}_I(t_2) \hat{V}_I(t_1) | i \rangle \propto & \langle f | \hat{\sigma}_2^\dagger(t_2) \hat{a}_\beta(t_2) \hat{\sigma}_1^\dagger(t_1) \hat{a}_\alpha(t_1) \\ & + \hat{\sigma}_1^\dagger(t_2) \hat{a}_\alpha(t_2) \hat{\sigma}_2^\dagger(t_1) \hat{a}_\beta(t_1) \\ & + (1 \leftrightarrow 2) | i \rangle. \end{aligned}$$

That is, there are four possible ways of achieving two-photon absorption: two *pathways* corresponding to which photon is absorbed first, and two *pairings* corresponding to which atom absorbs which photon, as shown in Fig. 1. Carrying out the nested time integrals now gives the Fermi golden rule in Eq. (2), and we find $A(\omega_1, \omega_2) \rightarrow 0$ due to destructive interference of excitation pathways, provided $\omega_1 - \omega_2 \neq \pm|\omega_\alpha - \omega_\beta|$.

The key point of this Letter is that the above interference does not take place when the photons are temporally entangled as in the cascade state $|\alpha, \beta\rangle$. In this case, we have a multimode, frequency-entangled state vector as given in Eq. (3). Following the monochromatic treatment above, we separate out $A(\omega_1, \omega_2)$ into four parts, depending on which atom absorbs which photon in which order:

$$A(\omega_1, \omega_2) = A_{\alpha\beta,12} + A_{\beta\alpha,21} + A_{\alpha\beta,21} + A_{\beta\alpha,12}, \quad (12)$$

where $A_{\mu\nu,ij}$ is defined as the probability amplitude that atom i absorbs photon μ first (time t_1), and atom j absorbs photon ν second (time t_2). This ‘‘time-ordered’’ excitation amplitude is closely related to the two-photon correlation amplitude $\psi_{\mu\nu}^{(2)}(\mathbf{r}_i, \mathbf{r}_j; t_1, t_2)$ in Eq. (5):

$$\begin{aligned} A_{\mu\nu,ij} = & \int_0^t dt_2 \int_0^{t_2} dt_1 \exp(i\omega_i t_1 + i\omega_j t_2) \\ & \times \psi_{\mu\nu}^{(2)}(\mathbf{r}_i, \mathbf{r}_j; t_1, t_2). \end{aligned} \quad (13)$$

The correspondence between excitation probability and two-photon correlation has been noted before for a single atom [16]. For two atoms, we have four contributions to the joint excitation amplitude $A(\omega_1, \omega_2)$, as listed in Eq. (12). The first two terms in this equation correspond to the atom-photon pairing $\alpha \leftrightarrow 1$ and $\beta \leftrightarrow 2$, while the last two terms correspond to the opposite pairing. For the cascade state, let us write out the terms corresponding to the absorption of α first, $A_{\alpha\beta,12}$ and $A_{\alpha\beta,21}$, by substituting the expression for $\psi_{\alpha\beta}^{(2)}$ into Eq. (13):

$$\begin{aligned} A_{\alpha\beta,ij} \propto & \int_0^t dt_2 \int_0^{t_2} dt_1 \exp(i\omega_i t_1 + i\omega_j t_2) \\ & \times \theta(\tau_\alpha) \exp[-(i\omega_\alpha + i\omega_\beta + \gamma_\alpha)\tau_\alpha] \\ & \times \theta(\tau_\beta - \tau_\alpha) \exp[-(i\omega_\beta + \gamma_\beta)(\tau_\beta - \tau_\alpha)]. \end{aligned} \quad (14)$$

This equation highlights the key difference between the absorption of classical and entangled light. In the classical (or separable) case, both excitation pathways shown in Fig. 1 are allowed, as either frequency ω_α or ω_β can be absorbed first [corresponding to interchanging t_1 and t_2 in

the integrand of Eq. (13)]. However, in the entangled case, a time ordering is imposed *at the source*, viz., $\theta(\tau_\beta - \tau_\alpha) = \theta[(t_2 - \Delta r_j) - (t_1 - \Delta r_i)]$. In particular, when the two atoms are equidistant from the source, $\Delta r_i = \Delta r_j$, the ordering of retarded times ($\tau_\beta > \tau_\alpha$) imposes a strict ordering of absolute times ($t_2 > t_1$) in the integrand. When combined with the limits of integration, this precludes terms $A_{\beta\alpha,21}$ and $A_{\beta\alpha,12}$, which correspond to absorption of β first. As a result, one of the two interfering pathways in each atom-photon pairing is always absent (dashed line in Fig. 1), and two-photon absorption becomes possible.

We evaluate the integral in Eq. (14) in the limit that $t - \Delta r_i \gg \gamma_\alpha^{-1}$ (which ensures that the photons have reached the atoms) and assume that the atoms are located much closer together than the distance c/γ_β . Then, the net two-atom excitation amplitude in Eq. (12) becomes, for the cascade state, $A(\omega_1, \omega_2) = A_{\alpha\beta,12} + A_{\alpha\beta,21}$, where

$$A_{\alpha\beta,12} = \frac{K}{(\omega_1 + \omega_2 - \omega_\alpha - \omega_\beta + i\gamma_\alpha)(\omega_2 - \omega_\beta + i\gamma_\beta)},$$

which contains a two-photon resonance term in the denominator, a departure from the classical Fermi golden rule result in Eq. (2). Furthermore, the excitation amplitude $A_{\alpha\beta,12}$ reproduces the spectral profile of the initial cascade state $|\alpha, \beta\rangle$, as can be seen by making the replacements $\omega_p \rightarrow \omega_1$ and $\omega_q \rightarrow \omega_2$ in Eq. (3).

Thus, time-frequency entanglement between the photons results in a *cooperative* absorption by the two atoms that enhances their joint excitation probability. We can make this a quantitative statement by relating it to the degree of photon entanglement in the cascade state. Working in the frequency domain, we define a normalized excitation probability $P_{\alpha\beta}(\omega_1, \omega_2) \propto |A_{\alpha\beta,12}|^2$, corresponding to the absorption of α by atom 1 and β by atom 2. Marginal single-atom probabilities $P_\alpha(\omega_1)$ and $P_\beta(\omega_2)$ may be calculated by integrating $P_{\alpha\beta}(\omega_1, \omega_2)$ over ω_2 and ω_1 , respectively [17]. We then find that the ratio of spectral probabilities of joint and separable absorptions for the cascade state is given by

$$\frac{P_{\alpha\beta}(\omega_1, \omega_2)}{P_\alpha(\omega_1)P_\beta(\omega_2)} = 1 + \frac{\gamma_\beta}{\gamma_\alpha} + \frac{(\omega_1 - \omega_\alpha)^2}{\gamma_\alpha(\gamma_\alpha + \gamma_\beta)}, \quad (15)$$

assuming exact two-photon resonance: $\omega_1 + \omega_2 = \omega_\alpha + \omega_\beta$. That is, we find that a joint excitation of both atoms together is more likely than individual excitations of the two atoms separately, and the gain in absorption rate is directly related to the degree of photon entanglement, $\gamma_\beta/\gamma_\alpha$, which is independent of one-photon detunings. In the limit $\gamma_\beta \gg \gamma_\alpha$, the two photons in the cascade state travel close together in the time domain [cf. discussion following Eqs. (7) and (8)], which favors a concurrent absorption over one that is uncorrelated in time.

To summarize, we have shown in this Letter that the joint excitation of two independent atoms is enhanced by

the time-frequency entanglement in the cascade two-photon state in two ways. First, from a qualitative standpoint, the two-photon absorption does not suffer from the destructive interference of excitation pathways that normally afflicts two-atom excitation, an advantage that owes to the time asymmetry in the entangled two-photon state which cannot be simulated classically. Second, from a quantitative standpoint, the enhancement in absorption rate is determined by the degree of photon entanglement created at the source, $\gamma_\beta/\gamma_\alpha$, which is related to the temporal proximity of the two photons. The results of this Letter may be generalized to the case of asymmetrically positioned atoms, $\Delta r_i \neq \Delta r_j$. Here one finds that the ordering of retarded times of photon propagation does not imply a strict ordering of absolute times of atomic excitation in Eq. (14), with the result that partial interference of excitation pathways is restored.

We believe our work suggests a new perspective on two-photon spectroscopy with entangled photons. Further, it also suggests possible applications to quantum information processes, as one might do entanglement transfer between photons and atoms using field-mode entanglement as a continuous variable resource.

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