Entanglement-Induced Two-Photon Transparency

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The rate of absorption of entangled photon pairs is linear in the photon-flux density. We demonstrate that the two-photon absorption cross section is a nonmonotonic function of the entanglement time; it vanishes for certain energy-level configurations and values of the entanglement time. This entanglement-induced two-photon transparency arises from the coherent summation of transition-amplitude contributions over the finite entanglement time. As an example, the entangled two-photon cross section for the $1S$-$2S$ electronic transition in atomic hydrogen is obtained. [S0031-9007(97)02508-8]

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Since the 1930s, when two-photon absorption was first described [1], multiphoton processes have received considerable attention as fundamental components of the interaction of light with matter. With classical light, the N-photon absorption and ionization rates vary with the photon-flux density ϕ as ϕ^N. These rates also depend on the statistical properties of the light. For example, it has been shown that the off-resonance rate using a thermal light source exceeds that using a coherent light source by a factor of $N!$ [2,3]. Current interests in classical-light induced multiphoton processes include two-photon fluorescence [4] and two-photon spectroscopy [5]. With the advent of nonclassical light sources [6,7], new phenomena in multiphoton processes can be explored. A linear (rather than quadratic) dependence of the two-photon absorption rate on photon-flux density has been predicted for sufficiently weak entangled [8,9] and quadrature squeezed light [10]; indeed, the latter has been recently observed with atomic cesium in a squeezed vacuum [11].

A composite quantum system whose state cannot be factored into a product of single-particle states is said to be entangled [7]; it has no classical analog. In this Letter, we present a quantum-mechanical calculation of the two-photon (linear) absorption rate with temporally entangled light. Our results reveal a new nonclassical phenomenon—nonmonotonic variations in the absorption rate as a function of the entanglement time. An important feature of these variations is the occurrence of significantly reduced values of the absorption cross section that emerge for certain parameter values, which we term *entanglement-induced two-photon transparency*. Like electromagnetically induced transparency [12,13], which has applications in lasing without inversion [13] and isotope discrimination [14], entanglement-induced transparency is a quantum interference effect. It is distinguished from the inhibition and enhancement of two-photon absorption using classical light [15] by its dependence on the degree of entanglement of the photon pair and its linear dependence on the photon-flux density. As an example, we calculate the effect in the $1S$-$2S$ transition of atomic hydrogen [16].

Simple probabilistic model.—We first present a simple probabilistic two-photon absorption model that considers the photons as particles. The process is regarded as having two steps: the first photon initiates a transition to a virtual state and the second photon brings about a transition to the final state. For randomly arriving photons, the probabilistic analysis yields a transition rate R_r that depends only on the material's single-photon cross section σ and virtual-state lifetime τ. The resulting random two-photon absorption rate is $R_r = \delta_r \phi^2$ where the two-photon quadratic cross section is $\delta_r = \sigma^2 \tau$ [17].

Next, consider correlated photon pairs arriving at the absorbing medium with flux density $\phi/2$ photon pairs/(cm^2 s). In this case, the absorption rate of the material depends on the probability $\xi(T_e)$ that the two photons emitted within the time T_e arrive within τ and the probability $\zeta(A_e)$ that the two photons emitted within the area A_e arrive within σ. Thus, the correlated two-photon absorption rate is $R_e = \sigma_e \phi$ with cross section $\sigma_e = \sigma \xi(T_e) \zeta(A_e)/2$. This rate must be supplemented by that representing the accidental arrival of pairs within τ and σ, resulting in the overall two-photon absorption rate

$$R = R_e + R_r = \sigma_e \phi + \delta_r \phi^2. \tag{1}$$

It is clear that correlated two-photon absorption dominates random two-photon absorption only when the photon-flux density is sufficiently small. The critical photon-flux density at which the two processes are equal is $\phi_c = \sigma_e / \delta_r$. For $T_e \ll \tau$ and $A_e \ll \sigma$, $\xi(T_e)$ and $\zeta(A_e)$ are unity, yielding $\sigma_e = \delta_r / 2\sigma\tau$. In the experimentally relevant case in which $T_e \gg \tau$ and $A_e \gg \sigma$, the probability functions are $\xi(T_e) = \tau / T_e$ and $\zeta(A_e) = \sigma / A_e$, yielding

$$\sigma_e = \delta_r / 2 A_e T_e \,. \tag{2}$$

Quantum-mechanical cross section.—We now obtain a proper quantum-mechanical expression for σ_e, which we then compare to the results obtained above. Entangled light is assumed to be created by parametric downconversion through a second-order nonlinear optical interaction [7,18]. This process produces an entangled photon pair (signal and idler) from a single pump photon such that energy and momentum are conserved. We consider collinear Type-II down-conversion in which the signal and idler photons have wave vectors parallel to that of the pump and have orthogonal polarizations [19]. Assuming a down-conversion crystal of length l, and a pump beam with wave number k_p, Gaussian spectrum of width $\Delta\omega_p$, and central frequency ω_p, the down-converted photon pair forms a time-entangled pure quantum state referred to as the twin state [19,20]

$$|\text{twin}\rangle = Nl \int\int d\omega_1\, d\omega_2 \exp\left[-\frac{(\omega_1 + \omega_2 - \omega_p)^2}{\Delta\omega_p^2}\right] \text{sinc}\left[\frac{l}{2\pi}(k_1 + k_2 - k_p)\right] |\omega_1, \omega_2\rangle . \tag{3}$$

Here ω_1, $k_1(\omega_1)$ and ω_2, $k_2(\omega_2)$ are the signal and idler frequencies and wave numbers, respectively. Typically, $\Delta\omega_p \ll \omega_p$ and $1/T_e \ll \omega_p$ [19,20] so that the signal and idler wave packets can be identified by their centers at ω_1^0, ω_2^0 and the normalization factor is given by $N^2 = T_e\sqrt{2/\pi^3}/l^2\Delta\omega_p$; units are chosen such that $\hbar = c = 1$. The entanglement time T_e, which is the temporal width of the fourth-order coherence function, is in this case equal to the difference between the mean transit times of the two photons. Because the signal and idler photons are orthogonally polarized and travel at slightly different group velocities u_1 and u_2, we have $T_e = T_1 - T_2 = l(1/u_1 - 1/u_2)/2$, where $u_1 < u_2$, and $T_1 = l/2u_1$ and $T_2 = l/2u_2$ are the *mean* transit times. In writing Eq. (3) we have assumed that the signal and idler have been externally delayed by a compensating time equal to $T_1 - T_2$. The use of the symbol T_e will be subsequently justified by its correspondence with that used in the simple probabilistic particle analysis.

We begin by considering the interaction of one entangled photon pair with a medium in an initial state $|\psi_i\rangle$ with energy eigenvalue ε_i. Thus, the initial state of the system is $|\Psi_i\rangle = |\psi_i\rangle \otimes |\text{twin}\rangle$. The excitation of the medium occurs through the intermediate states $|\psi_j\rangle$ with complex eigenvalues $\varepsilon_j + i\kappa_j/2$ [3]. The phenomenological intermediate state linewidths κ_j in general depend on the photon-flux density but can be considered constants for sufficiently weak light [21]. The final state of the light is the vacuum $|0,0\rangle$, and that of the material is $|\psi_f\rangle$ with eigenvalue ε_f, so that the final state of the system is $|\Psi_f\rangle = |\psi_f\rangle \otimes |0,0\rangle$. With this formulation, the absorption rate of the medium can be calculated in the interaction picture in a manner analogous to that used for two-photon absorption with classical light [21,22]. Using second-order time-dependent perturbation theory, the transition probability amplitude S_{fi} is

$$\begin{aligned} S_{fi} = {} & \frac{\pi N l}{2A_q}\sqrt{\omega_1^0\omega_2^0}\exp\left[-\frac{(\varepsilon_f - \varepsilon_i - \omega_p)^2}{\Delta\omega_p^2}\right] \\ & \times \sum_j \left\{ D_{21}^{(j)} \frac{1 - \exp\{-i[T_e(\varepsilon_j - \varepsilon_i - \omega_1^0) + (T_0 - T_e/2)(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0)] - T_e\kappa_j/2\}}{T_e(\varepsilon_j - \varepsilon_i - \omega_1^0) + (T_0 - T_e/2)(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0) - iT_e\kappa_j/2} \right. \\ & \left. + D_{12}^{(j)} \frac{1 - \exp\{-i[T_e(\varepsilon_j - \varepsilon_f + \omega_1^0) - (T_0 - T_e/2)(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0)] - T_e\kappa_j/2\}}{T_e(\varepsilon_j - \varepsilon_f + \omega_1^0) - (T_0 - T_e/2)(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0) - iT_e\kappa_j/2} \right\}, \end{aligned} \tag{4}$$

where $D_{kl}^{(j)} = \langle\psi_f|d_k|\psi_j\rangle\langle\psi_j|d_l|\psi_i\rangle$ are the transition matrix elements with material electric-dipole moment components d_k, d_l and $k, l = 1, 2$; $T_0 = (T_1 + T_2)/2$; and A_q is the quantization area. The sinc-function dependence of the entangled-two-photon wave function and the energy-level configuration of the medium combine to produce the complex dependence of S_{fi} on the nonlinear-material-dependent terms T_e and T_0, which characterize the structure of the entangled two-photon wave function.

We now generalize the interaction between the entangled photon pair and the medium to multiple pairs with an entanglement area A_e that arises from nonzero transverse momentum which is, however, sufficiently small so that Eq. (3) is still a valid description. Geometrical considerations analogous to those used in the simplified probabilistic analysis yield the absorption cross section for entangled light (the relatively small atomic size justifies a heuristic consideration of spatial effects):

$$\sigma_e = |S_{fi}|^2 A_q^2/A_e . \tag{5}$$

Equation (5), which is independent of A_q upon incorporation of Eq. (4), is our main result.

Special cases.—A deeper understanding of the physical nature of these results can be obtained by considering a simplified case in which the pump is monochromatic, so that $\Delta\omega_p \to 0$ and the phase matching condition $\omega_p = \omega_1^0 + \omega_2^0$ obtains. Under these conditions, we identify the energy mismatch $\Delta_k^{(j)} = \varepsilon_j - \varepsilon_i - \omega_k^0$ and Eqs. (4) and (5) reduce to

$$\begin{aligned} \sigma_e = {} & \frac{\pi}{4A_eT_e}\omega_1^0\omega_2^0\delta(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0) \\ & \times \left| \sum_j \left\{ D_{21}^{(j)} \frac{1 - \exp[-iT_e\Delta_1^{(j)} - T_e\kappa_j/2]}{\Delta_1^{(j)} - i\kappa_j/2} \right. \right. \\ & \left. \left. + D_{12}^{(j)} \frac{1 - \exp[-iT_e\Delta_2^{(j)} - T_e\kappa_j/2]}{\Delta_2^{(j)} - i\kappa_j/2} \right\} \right|^2 . \end{aligned} \tag{6}$$

Along with the inverse dependence on T_e, the structure of Eq. (6) permits constructive and destructive interference that can produce nonmonotonic behavior of σ_e as a function of T_e. Further simplification obtains for the special case of a medium in which a single intermediate state $j = s$ (with $T_e\kappa_s \approx 0$) dominates the summation so that we have a three-level system, and the entangled photons are degenerate with $\omega_1^0 = \omega_2^0 = \omega_p/2 \neq \varepsilon_s - \varepsilon_i$, whereupon

$$\sigma_e|_{j=s} = \frac{\pi[D_{21}^{(s)} + D_{12}^{(s)}]^2 \omega_p^2 \delta(\varepsilon_f - \varepsilon_i - \omega_p)}{4A_e T_e [\Delta_p^{(s)}]^2} \times \sin^2\left[\frac{\Delta_p^{(s)} T_e}{2}\right] \tag{7}$$

with $\Delta_p^{(s)} = (\varepsilon_s - \varepsilon_i - \omega_p/2)$. Full entanglement-induced two-photon transparency is then seen to occur at the zeros of the $\sin^2$ function: $T_e = 2m\pi/\Delta_p^{(s)}$, where $m = 1, 2, 3, \ldots.$

For comparison with Eq. (6), the quadratic two-photon absorption cross section for classical light is [22]

$$\delta_r = \frac{\pi}{2}\,\omega_1^0\omega_2^0\delta(\varepsilon_f - \varepsilon_i - \omega_1^0 - \omega_2^0) \times \left|\sum_j\left[\frac{D_{21}^{(j)}}{\Delta_1^{(j)} - i\kappa_j/2} + \frac{D_{12}^{(j)}}{\Delta_2^{(j)} - i\kappa_j/2}\right]\right|^2. \tag{8}$$

There are two significant differences between the entangled (linear) and the classical (quadratic) two-photon absorption cross sections. First, the entangled two-photon cross section contains a factor of $1/2A_eT_e$, which is just what is obtained for ϕ_c from the probabilistic particle analysis. Second, a T_e-dependent harmonic term appears that intertwines the parameters of the medium with the entangled-photon parameter in a manner that does not in general permit factorization. In the special case $\omega_1^0 = \omega_2^0 = \varepsilon_j - \varepsilon_i$ and $T_e\kappa_j \ll 1$, however, σ_e is maximized and Eq. (6) can be factored, giving $\sigma_e \propto T_e$ [9].

Origin of the interference.—In general, photon absorption results from the coherent summation of transition-amplitude contributions over all possible absorption times. The essential difference between the classical and entangled two-photon absorption cross sections arises because in the latter case the contributions are limited to photon pairs arriving within T_e [compare the summands in Eqs. (6) and (8)]. Although produced by entanglement in our study, we note that this type of quantum interference can occur in other situations involving sharp time windows. In all forms of two-photon absorption the contributions further depend on the order of the absorption, resulting in two terms within the summations in both Eqs. (6) and (8). These effects are in addition to the usual quantum interference that originates from the indistinguishability among amplitudes arising from different intermediate state transitions. (In electromagnetically induced transparency the interference is among the probability amplitudes of two or more transitions induced by two or more sources [12–14], whereas in entanglement-induced transparency the interference is among transition-amplitude contributions over the finite entanglement time window for a single transition induced by a single source.)

Our results are readily generalized to entangled N-photon absorption, which is expected to produce similar phenomena, including entanglement-induced N-photon transparency. Furthermore, it should also be possible to vary the absorption at particular signal and idler frequencies by manipulating the shape of the entangled-two-photon wave function, for example, by using multiple-crystal geometries.

Example: atomic hydrogen.—To explicitly demonstrate the nonmonotonicity, we consider entangled two-photon absorption in the exactly calculable case of atomic hydrogen. In particular, we evaluate the effect for the electronic $1S$-$2S$ transition which has been the focus of several recent experiments, including nonentangled two-photon absorption [5]. The selection rules of this transition forbid the absorption of two orthogonally polarized photons so that one of the photon beams is assumed to have had its polarization rotated to be parallel to that of the other beam. For simplicity, we consider a monochromatic source with $\omega_p = \varepsilon_f - \varepsilon_i$ and intermediate states with $\kappa_j = 0$ (since $\kappa_j \ll \Delta_k^{(j)}$ and $T_e\kappa_j \ll 1$). We take the $2S$ state to be Lorentzian broadened with a natural lifetime $1/\kappa_f = 122$ ms [5] and use $A_e = 10^{-6}$ cm^2. Numerical calculations of the entangled two-photon absorption rate are carried out using 50 principal quantum numbers (the use of additional intermediate states is insignificant on the outcome).

The results are shown in Figs. 1 and 2, for degenerate and nondegenerate entangled photon pairs, respectively [23]. These curves reveal large variations in the linear two-photon cross section over a range of

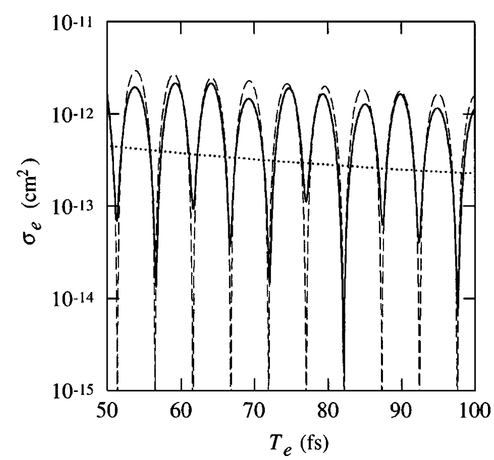

FIG. 1. Degenerate ($\omega_1^0 = \omega_2^0 = \omega_p/2$) entangled two-photon linear absorption cross sections for the $1S$-$2S$ transition in atomic hydrogen using Eq. (6) (solid curve), the single intermediate state approximation of Eq. (7) (dashed curve), and the result from the probabilistic particle analysis Eq. (2) with δ_r given by Eq. (8) (dotted curve).

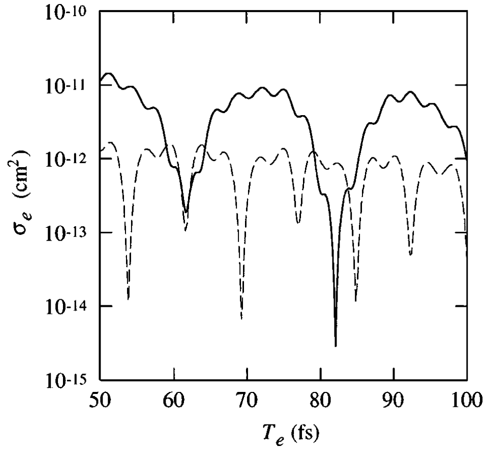

FIG. 2. Nondegenerate entangled two-photon linear absorption cross sections for the $1S$-$2S$ transition in atomic hydrogen using Eq. (6): $\omega_1^0 = \omega_p/3$, $\omega_2^0 = 2\omega_p/3$ (dashed curve); $\omega_1^0 = \omega_p/8$, $\omega_2^0 = 7\omega_p/8$ (solid curve).

entanglement times typically encountered in experiments [19]. The dominance of a single frequency in the degenerate curve of Fig. 1 is indicative of the contributions principally arising from the lowest intermediate state ($2P$). The cross section in this case (solid curve, Fig. 1) is well approximated by Eq. (7) (dashed curve, Fig. 1). The additional structure evident in the nondegenerate cases (Fig. 2) arises from the superposition of two different principal harmonic components, each of which is proportional to a different transition matrix element.

The minima in the cross sections lie 1 to 3 orders of magnitude below the maxima for both degenerate and nondegenerate absorption, demonstrating entanglement-induced transparency in a realistic physical system. Indeed, the modulation depth is nearly independent of T_e since typically $T_e \kappa_j < 10^{-5}$. This indicates that the linear two-photon absorption rate can be inhibited or enhanced by appropriate adjustment of the entanglement time over a wide range of values. This adjustment can be conveniently achieved through the use of a wedge-shaped crystal or the insertion of spectral filters in the signal and idler beams. However, for sufficiently large values of $T_e \kappa_j$, the factor $\exp(-T_e \kappa_j/2) \rightarrow 0$ in Eq. (6), whereupon the interference is washed out and the quantum-mechanical result coincides with the simple probabilistic particle formula: Eq. (2) with δ_r given by Eq. (8).

To observe these features in experiments, a sufficiently small sample or spot size may well be necessary to avoid a potential reduction of the modulation depth resulting from the range of optical path differences for different atoms in a three-dimensional sample. Preliminary studies involving spatiotemporal effects in entangled two-photon absorption indicate that the general features of our results persist over a wide range of nonideal conditions. The calculations presented here show that nonmonotonic absorption should occur in atomic hydrogen for $\phi < \phi_c \sim 10^{24}$ photons/(cm^2 s). Hence, the ease with which nonmonotonic entangled two-photon absorption can be attained is limited by the low efficiency of spontaneous down-conversion in nonlinear crystals ($\sim 10^{-7}$). This makes the production of large entangled-photon fluxes challenging and necessitates the use of state-of-the-art light sources and detectors, but it can be achieved.

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