Linear intensity dependence of a two-photon transition rate

Juha Javanainen and Phillip L. Gould
Department of Physics, University of Connecticut, Storrs, Connecticut 06269-3046
(Received 28 November 1988)

At low intensities the rate of two-photon transitions in an atom driven by parametrically down-converted light may be linear in intensity. Such a violation of the usual perturbative law arises from strong correlations between pairs of photons.

With recent advances in the generation and detection of antibunched,[1] sub-Poissonian,[2] and squeezed[3] light, the issue of how nonclassical radiation fields interact with atoms[4] becomes relevant. In this paper we consider a two-photon transition driven by parametrically down-converted light. Although such a transition is inherently a nonlinear process, we demonstrate a surprising result: at low intensity the high degree of photon correlations may lead to a transition rate which is linear in intensity.

In degenerate parametric down-conversion a nonlinear crystal transforms part of the incident light to light with twice the original wavelength, as if incident photons were split into pairs with half the energy. The down-converted light correspondingly exhibits an unusual degree of correlation, in that detection of a photon at one point in space-time may enormously enhance the probability of detecting its "twin" at another definite space-time location.[5] We shall assume that, by focusing or otherwise, the photons of a correlated pair are brought at the same time onto a three-level atom (Fig. 1) whose two-photon transition frequency nearly matches the frequency of the original light.

We may gain a qualitative understanding of the ensuing unusual behavior by contrasting the interaction of the three-level atom with ordinary coherent light and with light generated by parametric down-conversion. If the atom interacts with coherent light tuned to the two-photon resonance, the two-photon transition rate may be estimated as the rate of excitation from the (virtual or real) intermediate state times the probability that the atom is in the intermediate state. Since at low intensity both of these quantities are proportional to the intensity, we obtain the usual quadratic dependence of two-photon rate on intensity. On the other hand, with the parametrically down-converted light the excitation is accomplished in a single step: one photon of the pair promotes the atom to the virtual intermediate state, while its twin immediately (in a time less than the virtual-state lifetime) completes the two-photon transition. We may estimate the two-photon transition rate as the probability of excitation by a photon pair (independent of intensity) times the rate of arrival of photon pairs (proportional to intensity). The rate should be linear in intensity.

As a preamble to a more quantitative model we recall[6,7] the response of the three-level system (Fig. 1) to light in the Heisenberg picture using perturbation theory in the light intensity. The two well-known routes[8] from the ground state 0 to the excited state 2 can be isolated.

(i) One route is the two-step process, in which the field first promotes the atom to the intermediate state 1 and in the second step to the excited state 2. We mimic this sequence also in the calculations. When the field correlation time is much shorter than the inverse of the detunings, the two-step rate is given by

$$R_{\mathrm{TS}}(t)=4\left|\frac{d_{01}d_{12}}{\hbar^2}\right|\frac{1}{\Gamma}\left|\int_{-\infty}^{t}dt_1\langle\hat{E}^{-}(t)\hat{E}^{+}(t_1)\rangle\right|^2 . \tag{1}$$

Here d_{01} and d_{12} are the dipole-moment matrix elements of the transitions $0\to 1$ and $1\to 2$, and Γ is the decay rate of the population of state 1.

(ii) The other route is the two-photon process, which proceeds via the coherent superposition of states 0 and 2. The two-photon rate is

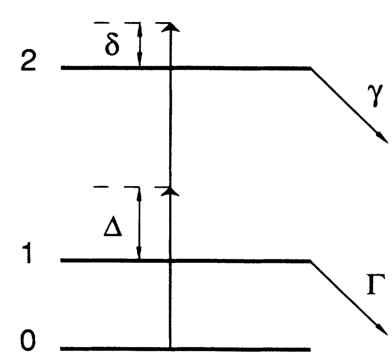

FIG. 1. Schematic of the three-level system under the two-photon excitation by down-converted light. The figure illustrates the two-photon (δ) and intermediate-state (Δ) detunings, and population decay rates of the intermediate state (Γ) and of the final state (γ).

$$R_{\mathrm{TP}}(t)=2\left|\frac{d_{01}d_{12}}{\hbar^2}\right|^2 \mathrm{Re}\left[\int_{-\infty}^{t} dt_1 e^{i(\delta-\Delta)(t-t_1)}\int_{-\infty}^{t_1} dt_2 e^{(i\delta-\gamma/2)(t_1-t_2)} \times\int_{-\infty}^{t_2} dt_3 e^{i\Delta(t_2-t_3)}\langle \hat{E}^-(t)\hat{E}^-(t_1)\hat{E}^+(t_2)\hat{E}^+(t_3)\rangle\right], \qquad (2)$$

where $\gamma/2$ is the decay rate of the superposition of the states 0 and 2, $\delta=2\omega-\omega_{21}$ is the detuning of the unsplit photons from two-photon resonance with the transition $0\rightarrow 2$, and $\Delta=\omega-\omega_{10}$ is the detuning from the intermediate-state resonance. $\hat{E}^{\pm}(\mathbf{r},t)$ are the positive- and negative-frequency components of the quantized electric field, in which the dominant time evolution $\exp(\mp i\omega t)$ at the average down-converted frequency ω has been factored out at the outset.

The most familiar four-field correlation function

$$G^{(2)}(\mathbf{r},t;\mathbf{r}',t')=\langle \hat{E}^-(\mathbf{r},t)\hat{E}^-(\mathbf{r}',t')\hat{E}^+(\mathbf{r}',t')\hat{E}^+(\mathbf{r},t)\rangle , \qquad (3)$$

essentially the joint probability that two detectors placed at $\mathbf{r}$ and $\mathbf{r}'$ will record photon counts at times t and t',[9] does not always suffice to predict the two-photon transition rate. In fact, we need a more general correlation function. As even $G^{(2)}$ already is a highly nontrivial object for spontaneously down-converted light,[10–12] we shall resort to heuristic modeling of the field correlations.

First, the spectrum of down-converted light may be quite broad.[13] Accordingly, the Fourier transform of the spectrum, i.e., the two-field correlation function $\langle \hat{E}^-(\mathbf{r},t_1)\hat{E}^+(\mathbf{r},t_2)\rangle$,[9] decays from its value E^2 at $t_1=t_2$ on a time scale $|t_1-t_2|\sim\tau$ that is henceforth taken to be the shortest time scale of the problem. We write a qualitative ansatz valid inside time integrals,

$$\langle \hat{E}^-(\mathbf{r},t_1)\hat{E}^+(\mathbf{r},t_2)\rangle=\tau E^2\delta(t_1-t_2) . \qquad (4)$$

Next, the photon density of the electromagnetic field with intensity $I=c\epsilon_0E^2/2$ is conveniently defined as $\epsilon_0E^2/2\hbar\omega$. We may thus regard $(\epsilon_0/2\hbar\omega)^2G^{(2)}$ as the joint probability density for two photons. Corresponding to the notion of a photon pair, we define a coherence area A in the plane perpendicular to the direction of propagation of light and a longitudinal coherence time τ_c such that if one photon is detected at $(\mathbf{r},t)$, then its twin resides in a volume $Ac\tau_c$ surrounding $(\mathbf{r},t)$. In terms of one- and two-photon densities, we have the estimate[12,14]

$$\int dS'\,c\,dt'\left[\frac{\epsilon_0}{2\hbar\omega}\right]^2 \times\langle \hat{E}^-(\mathbf{r},t)\hat{E}^-(\mathbf{r}',t')\hat{E}^+(\mathbf{r}',t')\hat{E}^+(\mathbf{r},t)\rangle=\frac{\epsilon_0}{2\hbar\omega}E^2 , \qquad (5)$$

where the integrand is assumed effectively nonzero over the coherence time and area only. Since the four-field correlation function inside the integral falls off at the temporal and spatial scales $|t-t'|\sim\tau_c$, $|\mathbf{r}-\mathbf{r}'|\sim A^{1/2}$, it should have a maximum of the order $(E^2\hbar\omega)/(c\epsilon_0A\tau_c)$ at $\mathbf{r}=\mathbf{r}'$, $t=t'$. Accordingly, we set up the following "top hat" model for $G^{(2)}$:

$$G^{(2)}(\mathbf{r},t;\mathbf{r}',t')=\begin{cases}\dfrac{2E^2\hbar\omega}{c\epsilon_0A\tau_c} & \text{for } |\mathbf{r}-\mathbf{r}'|<\sqrt{A/\pi},\ |t-t'|<\tau_c/2\\ 0 & \text{otherwise .}\end{cases} \qquad (6)$$

The final element required to characterize the light field is the factorization of the four-field correlation function into a product of two-photon amplitudes, valid in the neighborhood of the peak,[10]

$$\langle \hat{E}^-(\mathbf{r},t_1)\hat{E}^-(\mathbf{r}_2,t_2)\hat{E}^+(\mathbf{r}_3,t_3)\hat{E}^+(\mathbf{r}_4,t_4)\rangle =\langle \hat{E}^-(\mathbf{r}_1,t_1)\hat{E}^-(\mathbf{r}_2,t_2)\rangle\langle \hat{E}^+(\mathbf{r}_3,t_3)\hat{E}^+(\mathbf{r}_4,t_4)\rangle , \qquad (7)$$

Comparison of (3), (6), and (7) now fixes the form of the two-photon amplitude to within a position- and time-dependent phase factor. With the further assumption that the phase does not vary in time on a scale shorter than τ_c, we obtain a model analogous to (4) for the two-photon amplitude,

$$\langle \hat{E}^+(\mathbf{r},t_1)\hat{E}^+(\mathbf{r},t_2)\rangle=\begin{cases}e^{i\phi(\mathbf{r})}\left[\dfrac{2E^2\hbar\omega}{\epsilon_0cA\tau_c}\right]^{1/2} & \text{for } |t_1-t_2|<\tau_c/2\\ 0 & \text{otherwise}\end{cases} =e^{i\phi(\mathbf{r})}\left[\frac{2E^2\hbar\omega\tau_c}{\epsilon_0cA}\right]^{1/2}\delta(t_1-t_2) . \qquad (8)$$

Here we assume that τ_c is a short time scale relative to the atomic response. ϕ is a phase that will shortly cancel.

Inserting (4), (7), and (8) into (1) and (2), we obtain the total transition rate as a sum of two-step and two-photon contributions,

$$R=\left|\frac{d_{01}d_{12}}{\hbar^2}\right|^2\left[\frac{E^4\tau^2}{\Gamma}+\frac{\gamma/2}{\delta^2+(\gamma/2)^2}\frac{E^2\hbar\omega\tau}{\epsilon_0 c A}\right]. \quad (9)$$

The intermediate-state detuning Δ is absent from (9), as expected from our assumption that $\Delta\tau \ll 1$. Our argument was based on the top hat shape (6) for $G^{(2)}$, arguably the simplest quantitative model of the experimental fact that two detectors record an essentially perfect correlation of photon counts when placed in appropriate field positions. Other choices such as a Gaussian shape in space and time of $G^{(2)}$ would have simply altered the numerical factors in (9). Here and below we choose the one- and two-photon correlation times equal, $\tau=\tau_c$, i.e., we assume that τ_c is determined by the bandwidth of the down-converted light.[12,13]

According to (9), at low intensities the two-photon rate linear in I dominates. If the decay rates Γ and γ are equal, at exact two-photon resonance ($\delta=0$) the crossover to the conventional I^2 law occurs at $I_c=\hbar\omega/\tau A$. This condition corresponds to a photon density such that two uncorrelated photons are likely to be found inside a coherence volume.

There are two criteria which must be met in order to experimentally observe the effect as we have described it. First, we require an appropriate two-photon transition, i.e., one with a nearly resonant intermediate state. For a correlation time of 100 fs,[5] an intermediate state detuning of less than 50 cm^{-1} is necessary. The second criterion involves the two-photon rate in the linear regime. The relevant figure of merit is the transition rate at the crossover intensity I_c. If we again assume two-photon resonance ($\delta=0$) and take the two decay rates γ and Γ to be equal, then the linear rate at I_c is

$$R_c=\frac{1}{\gamma}\left[\frac{4\pi d_{01}d_{12}}{\hbar\lambda\epsilon_0 A}\right]^2. \quad (10)$$

For a numerical example we consider collinear phase matching at degeneracy,[13] in which case the original and the down-converted light essentially copropagate. It seems reasonable to assume that the coherence area A is determined solely by the subsequent focusing of the down-converted light, i.e., A equals the area of the focus.[11] Suppose that the down-converted light is focused to a Gaussian waist w_0 ($1/e^2$ radius of intensity) giving $A=\pi w_0^2$, then the focal volume is given by the area times twice the Rayleigh range,

$$V=A\left[2\frac{\pi w_0^2}{\lambda}\right]=2\frac{A^2}{\lambda}. \quad (11)$$

Illuminating a gas of atoms at density n (assuming all atoms are two-photon resonant) which have the generic matrix elements $d_{01}=d_{12}=ea_0$ (a_0 is the Bohr radius) and the decay rates

$$\gamma=\Gamma=\frac{d^2}{3\pi\epsilon_0\hbar}\left[\frac{2\pi}{\lambda}\right]^3, \quad (12)$$

we expect a total number of transitions per unit time within the focal volume

$$R_c nV=48\pi\alpha c a_0^2 n=48\pi\frac{E_H}{\hbar}(a_0^3 n), \quad (13)$$

where α denotes the fine-structure constant and E_H is the Hartree energy. Quite surprisingly, if the focus sets the coherence area, the maximum achievable linear excitation rate of the entire atomic sample (atoms per second) is independent of the focal volume: a smaller focal volume is compensated for by more transitions per atom.[15] For a density of $n=10^{12}$ cm^{-3}, the rate is approximately 10^6 s^{-1}, and should be readily observable using photoionization detection.

Our example produces another surprise, too: the down-converted power corresponding to the crossover intensity $I_c A$ varies inversely with τ, but is *independent* of A and thus of focusing. For light of wavelength $\lambda=1$ μm and correlation time $\tau=100$ fs, we require approximately 2×10^{-6} W. Parametric oscillators[16] can provide the required power, but their large coherence times are likely to prove prohibitive. Pulsed experiments[13,17] have demonstrated correlation times as short as 200 fs and peak powers as high as 1 MW. Moreover, a conversion efficiency approaching 10^{-2} may be obtained via parametric amplification following the spontaneous parametric emission, and the correlations between twin photons are preserved in the process.[13] Unfortunately, the small duty cycle of pulsed experiments degrades the effective total transition rate. As it comes to continuous-wave experiments, nonlinear crystals can spontaneously down-convert with typical efficiencies of 10^{-7}. Hence about 10 W of pumping light is needed to generate 1 μW of down-converted power.

The special importance of the statistical properties of the light used to drive a two-photon transition is, of course, well known.[18] For instance, the transition rate for chaotic light is twice that for coherent light; larger intensity fluctuations simply give a larger mean value of the square of the intensity. The dependence of the transition rate on average intensity remains quadratic, however. An experiment to demonstrate the linear intensity dependence of a two-photon transition rate in parametrically down-converted light would at this writing be the first observation of qualitatively new physics resulting from quantum correlations in the light driving atomic transitions. Moreover, such an experiment appears feasible with the present state-of-the-art technology.

Note added. After the present paper was originally submitted, Julio Gea-Banacloche[19] pointed out that the two-photon absorption rate in a low-intensity squeezed vacuum is linear in intensity. As the squeezed vacuum is a superposition of even photon number states and the photons therefore occur in pairs, his result evidently is closely related to ours.

We are grateful to L. Mandel for his opinion on the status of the analytical theory of down-conversion, which has substantially shaped this work. Helpful discussions with A. Aspect, R. Chiao, H. J. Kimble, and P. Zoller are also acknowledged. This work is partially supported by the National Science Foundation, Grants No. PHY-8857336 and No. PHY-8902659.

[1]H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. A **18**, 201 (1978).

[2]R. Short and L. Mandel, Phys. Rev. Lett. **51**, 384 (1983).

[3]See, for example, the following special issues on squeezed states: J. Opt. Soc. Am. B **4** (10) (1987); and J. Mod. Opt. **34** (6/7) (1987).

[4]P. L. Knight and D. T. Pegg, J. Phys. B **15**, 3211 (1982); C. W. Gardiner, Phys. Rev. Lett. **56**, 1917 (1986); J. Janszky and Y. Yushin, Phys. Rev. A **36**, 1288 (1987); H. Ritsch and P. Zoller, Opt. Commun. **64**, 523 (1987); H. J. Carmichael, A. S. Lane, and D. F. Walls, Phys. Rev. Lett. **58**, 2639 (1987); A. S. Lane, M. D. Reid, and D. F. Walls, *ibid.* **60**, 1940 (1988).

[5]D. Burnham and D. Weinberg, Phys. Rev. Lett. **25**, 84 (1970); C. K. Hong, Z. Y. Ou, and L. Mandel, *ibid.* **59**, 2044 (1987).

[6]G. S. Agarwal, Phys. Rev. A **1**, 1445 (1970); J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D **10**, 3350 (1974); H. J. Kimble and L. Mandel, Phys. Rev. A **13**, 2123 (1976).

[7]The additional technical assumption that the damping of the three-state system (γ and Γ) either is of nonelectromagnetic origin or results from radiation reaction acting back on transitions other than $0\rightarrow 1$ and $1\rightarrow 2$ is helpful here, as it allows one to commute freely the atomic operators and the operators of the incident field. For convenience we write the relevant decay rate of the 0-2 superposition as $\gamma/2$, as if γ were the population decay rate from the excited state 2.

[8]See, e.g., S. Stenholm, *Foundations of Laser Spectroscopy* (Wiley, New York, 1984).

[9]R. J. Glauber, Phys. Rev. **130**, 2529 (1963); **131**, 2766 (1963).

[10]B. R. Mollow, Phys. Rev. A **8**, 2684 (1973).

[11]D. N. Klyshko, Zh. Eksp. Teor. Fiz. **83**, 1313 (1982) [Sov. Phys.—JETP **56**, 753 (1982)]; **94**, 82 (1988) [**67**, 1131 (1988)].

[12]C. K. Hong and L. Mandel, Phys. Rev. A **31**, 2409 (1985).

[13]I. Abram, R. K. Raj, J. L. Oudar, and G. Dolique, Phys. Rev. Lett. **57**, 2516 (1986).

[14]This is related to the equality of one- and two-photon detection probabilities discussed in detail in Ref. 12 [see Eq. (46)].

[15]This is true for a two-photon transition driven by ordinary coherent light as well. However, in that case the dependence on laser power is quadratic, while in our case it is linear.

[16]A. Heidmann, R. J. Horowicz, S. Reynaud, E. Giacobino, and C. Fabre, Phys. Rev. Lett. **59**, 2555 (1987); S. Reynaud, C. Fabre, and E. Giacobino, J. Opt. Soc. Am. B **4**, 1520 (1987); M. D. Reid and P. D. Drummond, Phys. Rev. Lett. **60**, 2731 (1988).

[17]R. E. Slusher, P. Grangier, A. LaPorta, B. Yurke, and M. J. Potasek, Phys. Rev. Lett. **59**, 2566 (1987).

[18]M. W. Hamilton, K. Arnett, S. J. Smith, D. S. Elliott, M. Dziemballa, and P. Zoller, Phys. Rev. A **36**, 178 (1987).

[19]J. Gea-Banacloche, Phys. Rev. Lett. **62**, 1603 (1989).