Generation of Pair Coherent States and Squeezing via the Competition of Four-Wave Mixing and Amplified Spontaneous Emission

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Strong competition between four-wave mixing and amplified spontaneous emission in resonant two-photon excitations is shown to generate radiation fields with strong squeezing and antibunching. The generated fields are in a new type of coherent state which is an eigenstate of the operator corresponding to the simultaneous annihilation of photons in two modes.

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In this Letter I report the generation of fields with strong quantum features such as squeezing and antibunching.[1] I examine the recent experimental work[2] of Malcuit, Gauthier, and Boyd, who reported the suppression of amplified spontaneous emission (ASE) in the two-photon resonant excitation of the sodium $3d$ level. In their experiment the suppression occurs as a result of the competition between the four-wave mixing (FWM) processes and ASE. The generated modes grow from vacuum and therefore the quantum features of the radiation field should be important. Hence, in order to understand the nature of the generated fields, a fully quantized treatment of the system studied by Malcuit, Gauthier, and Boyd is warranted. I formulate a quantum statistical theory of this experiment. The theory will also be applicable to related systems involving resonant two-photon excitations. I present an exact solution for the density matrix of the generated field. I demonstrate that the generated field is in a new type of coherent state which I refer to as the pair coherent state. The pair coherent state is an eigenstate of the operator corresponding to the simultaneous annihilation of photons in two modes. This pair coherent state is distinct[3] from the other known coherent states such as two-photon coherent states, atomic coherent states, or the SU(1,1) coherent states. I present explicit results for squeezing and antibunching in the generated fields for a range of system parameters involving the susceptibilities for FWM and two-photon absorption. I predict quite significant squeezing in the generated fields.

In the experiment of Malcuit, Gauthier, and Boyd, a pump laser of frequency ω_1 is used for the two-photon excitation of the Na $3d$ level ($|1\rangle$) starting from the ground state $3s$ ($|3\rangle$). The pump laser is assumed to be sufficiently detuned from the intermediate level $3p$ ($|2\rangle$). As a result of the four-wave mixing process two radiation fields of frequencies ω_2 and ω_3 are generated. The amplified spontaneous emission is proportional to the excited state ($3d$) population. The generated photons at ω_2 and ω_3 can be reabsorbed by a two-photon absorption process. I treat the pump field $E(\omega_1)$ classically and ignore its depletion. The generated fields at ω_2 and ω_3 are treated quantum mechanically. Let $b, b^\dagger$ ($c, c^\dagger$) be the annihilation and creation operators for the field at ω_2 (ω_3). I start from a microscopic Hamiltonian describing the interaction of a three-level atomic system with three radiation fields at ω_1, ω_2, and ω_3. The amplified spontaneous emission from the system is described by use of the usual master equation techniques[4]; i.e., the relaxation parameters in the equations of motion for the atomic system have contributions from spontaneous emission. I assume that the pump is detuned sufficiently enough so that the intermediate state $|2\rangle$ does not get populated. This permits me to ignore the spontaneous emission from the intermediate state. The spontaneous emission from the state $|1\rangle$ is included through the relaxation parameter Γ_2 [Eq. (5)]. The pump is assumed to be unidirectional. In this work I also ignore the saturation effects. This leads to considerable simplification which is in contrast to the single-photon resonant situations.[5] Using the adiabatic elimination of the atomic variables, and assuming the resonant condition $2\omega_1 = \omega_{13} = \omega_2 + \omega_3$, I have proved that the dynamical evolution of the fields at ω_2 and ω_3 is described by the master equation for the field density matrix ρ,

$$\partial\rho/\partial t = -i[H_{\text{eff}}, \rho] - \tfrac{1}{2}\kappa(b^\dagger c^\dagger bc\rho - 2bc\rho b^\dagger c^\dagger + \rho b^\dagger c^\dagger bc). \tag{1}$$

Here H_{eff} describes the four-wave mixing process[5]

$$H_{\text{eff}} = Gb^\dagger c^\dagger + \text{H.c.} \tag{2}$$

The coupling constant G is proportional to $E^2(\omega_1)$ and the susceptibility[6] $\chi^{(3)}(\omega_1, \omega_1, \omega_2)$ for four-wave mixing:

$$G = -2\pi\omega_3\chi^{(3)}(\omega_1, \omega_1, \omega_2)E^2(\omega_1) = -2\pi\omega_2\chi^{(3)}(\omega_1, \omega_1, \omega_3)E^2(\omega_1). \tag{3}$$

The parameter κ is related to the susceptibility for two-photon absorption[6] $\chi^{(3)}(\omega_3, -\omega_3, \omega_2)$ through

$$\kappa = 8\pi^2\omega_2\omega_3\hbar \, \mathrm{Im}\chi^{(3)}(\omega_3, -\omega_3, \omega_2)/V, \tag{4}$$

where V is the quantization volume for the field mode. The susceptibilities for the three-level atomic model are well known (cf. Ref. 2):

$$\chi^{(3)}(\omega_1, \omega_1, \omega_3) = \frac{\Delta_3}{\Delta_1}\chi^{(3)}(\omega_3, -\omega_3, \omega_2) = \frac{iN|d_{12}|^2|d_{23}|^2}{\hbar^3\Delta_1\Delta_3\Gamma_2}, \tag{5}$$

where

$$\Delta_1 = \omega_{23} - \omega_1, \quad \Delta_3 = \omega_{23} - (2\omega_1 - \omega_2), \tag{6}$$

Γ_2 is the linewidth of the two-photon transition, and N is the atomic number density.

Solution of the field density matrix equation (1) will yield all the statistical information on the quantum-mechanical generation of the fields ω_2 and ω_3. It may be added that solutions of (1) in the limiting cases (a) $\kappa = 0$ and (b) $G = 0$ are known[7] in different physical contexts. In the present system both processes are important and we would like to know the solution for arbitrary values of G/κ. Note that Eq. (1) admits an important conservation law,

$$\langle (b^\dagger b - c^\dagger c)p \rangle = 0, \quad p = 1, 2, \ldots . \tag{7}$$

This conservation law is useful in finding the solution of (1). The mean-value equations for the field amplitudes are

$$\langle \dot{b} \rangle = -iG\langle c^\dagger \rangle - \tfrac{1}{2}\kappa\langle bc^\dagger c \rangle, \quad \langle \dot{c} \rangle = -iG\langle b^\dagger \rangle - \tfrac{1}{2}\kappa\langle cb^\dagger b \rangle, \tag{8}$$

and thus an infinite hierarchy of equations is generated. This hierarchy could be closed by making suitable approximation; for example, in the semiclassical limit $\langle bc^\dagger c \rangle \sim \langle b \rangle |\langle c \rangle|^2$, I recover the equations of Ref. 2.

The suppression effects are dominant in the limit of long samples or in the steady-state limit.[2,8] In this limit I have found a complete quantum-mechanical solution to the density matrix. Note that the fields ω_2 and ω_3 grow from quantum noise and therefore in view of the conservation law (7), I look for a solution of the form

$$\rho = \sum_{m,n=0}^{\infty} \beta_{mn} |m,m\rangle\langle n,n|. \tag{9}$$

The coefficients β_{mn} are found from the solution of the recursion relation

$$\tfrac{1}{2}\kappa[\beta_{mn}(m^2+n^2) - 2(m+1)(n+1)\beta_{m+1,n+1}] + iG\beta_{m,n+1}(n+1) + iG^*\beta_{m,n-1}n - iG^*\beta_{m+1,n}(m+1) - iG\beta_{m-1,n}m = 0. \tag{10}$$

I have proved that Eq. (10) possesses the solution

$$\beta_{mn} = \frac{\zeta^m\zeta^{*n}}{m!n!}\beta_{00}, \quad \zeta = -\frac{2iG}{\kappa}, \tag{11}$$

where β_{00} can be obtained from the normalization of the density matrix. On combining (9) and (11) I get one of the key results of this Letter: The steady-state field density matrix is

$$\rho = |\zeta\rangle_{pp}\langle\zeta|, \quad \zeta = -2iG/\kappa, \tag{12}$$

$$|\zeta\rangle_p = N_0\sum_m \frac{\zeta^m}{m!}|m,m\rangle, \quad N_0^2\sum_m \frac{|\zeta|^{2m}}{m!^2} = 1, \tag{13}$$

where N_0 is the normalization constant. It can be checked that the state $|\zeta\rangle_p$ is an eigenstate of the operator bc:

$$bc|\zeta\rangle_p = \zeta|\zeta\rangle_p. \tag{14}$$

The operator bc is the pair annihilation operator, i.e., it corresponds to the simultaneous annihilation of photons of frequencies ω_2 and ω_3. Thus $|\zeta\rangle_p$ is a new type of coherent state associated with the pair annihilation operator. The state $|\zeta\rangle_p$ can be expressed in terms of the usual oscillator coherent states[3] $|z\rangle_b, |z\rangle_c$ for the two modes,

$$|\zeta\rangle_p = \int \frac{d\theta}{2\pi}|\sqrt{\zeta}e^{i\theta}\rangle_b|\sqrt{\zeta}e^{-i\theta}\rangle_c N_0 e^{|\zeta|}. \tag{15}$$

Note that the state $|\zeta\rangle_p$ is *not* the same as the two-photon coherent states discussed recently in the literature.[3] In view of the symmetry with respect to b and c, we can identify $|\zeta|^{1/2}$ as the steady-state value of the semiclassical field amplitude. Thus the steady-state value of the field intensity in semiclassical approximation will be $|\zeta|$, the explicit form of which can be obtained from Eqs. (3)–(5).

Having obtained the complete density matrix for the generated fields, I can now examine the quantum features of the generated field. I show in Fig. 1 the mean photon number $n=\langle b^\dagger b\rangle=\langle c^\dagger c\rangle$, in each mode, as a function of the parameter $|\zeta|$. I also exhibit its deviation $|\zeta|-n$ from the semiclassical value $|\zeta|$. The antibunching properties of the generated field are characterized in terms of $g^{(2)}=(\langle b^{\dagger 2}b^2\rangle-\langle b^\dagger b\rangle^2)/\langle b^\dagger b\rangle^2$ which is also shown in Fig. 1. This figure exhibits the very striking quantum nature of the generated field.

I next examine the squeezing characteristics of the output radiation. For this purpose it is sufficient[9] to find the variance in the operator

$$d=e^{-i\phi/2}(b+c)/2\sqrt{2}+\text{H.c.} \tag{16}$$

Here ϕ is the phase of the local oscillator used to mix the output fields. Using Eq. (12) I find

$$\langle :d^2:\rangle\equiv-\tfrac{1}{2}S=\tfrac{1}{2}\{n+\tfrac{1}{2}(\zeta e^{-i\phi}+\zeta^* e^{i\phi})\}. \tag{17}$$

Assuming that $\zeta=|\zeta|e^{i\theta}$, it is evident that squeezing is obtained if $n+|\zeta|\cos(\phi-\theta)<0$, i.e., if $\cos(\phi-\theta)<0$ or $\pi/2<(\phi-\theta)<\pi$. Maximum squeezing is obtained when $\phi-\theta=\pi$. In Fig. 1 I have shown $S=|\zeta|-n$, if $\phi-\theta=\pi$, as a function of $|\zeta|$. It is clear from the figure that one gets considerable squeezing (e.g., the variance $\langle d^2\rangle$ is 50% of the coherent-state value over a wide range of ζ values) in the radiation generated via the competition of ASE and FWM in resonant two-photon excitations. It is also interesting to note that the squeezing and the departure of the photon number from the semiclassical result coincide in this case. Thus, a good amount of squeezing should be observable in resonant two-photon excitations in vapors like Na and Rb in the limit of long samples by changing the parameter ζ which can be varied over a wide range by changes in the intensity and frequency of the pump field as is evident from Eqs. (3)–(5).

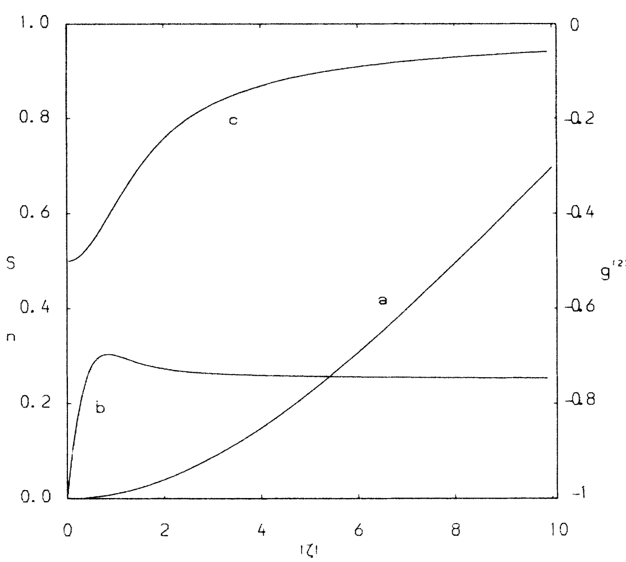

FIG. 1. The mean photon number n (curve a), its deviation $|\zeta|-n$ from the semiclassical result, squeezing parameter $S(=|\zeta|-n)$ (curve b), and $g^{(2)}$ (curve c) all as functions of the parameter $|\zeta|$. Note that $|\zeta|$ is related to the ratio of the susceptibilities for four-wave mixing and two-photon absorption. The scale for curve c is displayed on the right-hand side. The actual scale on the x axis for curve a is one tenth of that shown.

Finally, the amplified spontaneous emission is proportional to the population in the state $|1\rangle$. The atomic population in the state $|1\rangle$ can be obtained by considering an effective two-photon Hamiltonian

$$H_{\text{eff}}=[M_pE^2(\omega_1)+M_G bc]|1\rangle\langle 3|+\text{H.c.} \tag{18}$$

Here M_p (M_G) is the effective matrix element corresponding to the absorption of two photons of the pump (generated fields). These matrix elements are related to $\chi^{(3)}(\omega_1,-\omega_1,\omega_1)$ and $\chi^{(3)}(\omega_2,-\omega_2,\omega_3)$. Using Eq. (5) and the solution (12) for the field density matrix, I have shown that the effective quantum-mechanical field responsible for creating population in the state $|1\rangle$ is such that all its normally ordered moments are zero, i.e.,

$$\langle [M_pE^2(\omega_1)+M_G bc]^{\dagger p}[M_pE^2(\omega_1)+M_G bc]^q\rangle=0, \tag{19}$$

where p and q are positive integers. Note that (19) is the property of the vacuum of a quantum-mechanical field. Therefore, the effective interaction (18) involves interaction with an effective field with property (19) and hence no population is produced in the state $|1\rangle$; i.e., ASE is suppressed, which is in accordance with the experiment.[2]

In conclusion, I have shown how the strong competition between four-wave mixing and amplified spontaneous emission in resonant two-photon excitations produces radiation fields with striking quantum properties like antibunching and squeezing. This system produces new types of coherent states of the radiation fields involving annihilation of photons in pairs.

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Note added.— Since this paper was submitted I learned, from Dr. J. R. Klauber and Dr. S. M. Barnett to whom I am grateful, that the pair coherent states have been encountered before in connection with the Abelian charge, etc.[10]

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[1]See, for example, D. F. Walls, Nature (London) **306**, 141 (1983); C. K. Hong and L. Mandel, Phys. Rev. A **32**, 974 (1985); P. Kumar and J. H. Shapiro, Phys. Rev. A **30**, 1568 (1985), and references therein.

[2]M. S. Malcuit, D. J. Gauthier, and R. W. Boyd, Phys. Rev. Lett. **55**, 1086 (1985).

[3]R. J. Glauber, Phys. Rev. **131**, 2766 (1963); H. P. Yuen, Phys. Rev. A **13**, 2226 (1976); C. M. Caves and B. L. Schumaker, Phys. Rev. A **31**, 3068, 3093 (1985); A. M. Perelomov, Usp. Fiz. Nauk **123**, 23 (1977) [Sov. Phys. Uspekhi **20**, 703 (1977)].

[4]G. S. Agarwal, in *Quantum Optics,* edited by G. Höhler, Springer Tracts in Modern Physics Vol. 70 (Springer, Berlin, 1974).

[5]Several recent works [cf. M. D. Reid and D. F. Walls, Phys. Rev. A **32**, 396 (1985); M. Sargent, III, D. A. Holm, and M. S. Zubairy, Phys. Rev. A **31**, 3112 (1985)] discuss the quantum theory of four-wave mixing.

[6]I deal only with the lowest-order nonlinearities described by $\chi^{(3)}$. This is sufficient for most experiments including that of Ref. 2. Considerations of higher-order nonlinearities resulting, say, from $\chi^{(5)}(\omega_3, -\omega_3, \omega_2, -\omega_2, \omega_2)$ will lead to a much more complex master equation.

[7]B. R. Mollow and R. J. Glauber, Phys. Rev. **160**, 1076 (1967); H. D. Simaan and R. Loudon, J. Phys. A **8**, 539, 1140 (1975).

[8]J. C. Miller and R. N. Compton, Phys. Rev. A **25**, 2056 (1982); M. G. Payne and W. R. Garrett, Phys. Rev. A **26**, 356 (1982); D. J. Jackson and J. J. Wynne, Phys. Rev. Lett. **49**, 543 (1982); G. S. Agarwal and S. P. Tewari, Phys. Rev. A **29**, 1922 (1984).

[9]M. D. Levenson, R. M. Shelby, A. Aspect, M. Reid, and D. F. Walls, Phys. Rev. A **32**, 1550 (1985).

[10]J. R. Klauder and B. S. Skagerstam, *Coherent States* (World Scientific, Singapore, 1985), p. 43.