Single-Mode Generation of Quantum Photon States by Excited Single Molecules in a Microcavity Trap

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(Received 2 June 1995)

The active microcavity is adopted as an efficient source of nonclassical light. Using this device, excited with a mode-locked laser at a rate of 100 MHz, single photons are generated over a *single* field mode with a nonclassical sub-Poissonian distribution. Adiabatic recycling within a multistep Franck-Condon molecular optical-pumping mechanism, characterized by quantum efficiency close to 1, implies a pump self-regularization process leading to a striking n-squeezing effect. Replicating the basic single-atom excitation process a beam of quantum photon $|n\rangle$ states (Fock states) can be created. This represents a significant advance in the fields of basic quantum-mechanical investigation, quantum communication, and quantum cryptography.

PACS numbers: 42.50.Dv, 32.50.+d, 42.50.Vk

The generation of nonclassical light is an important topic of modern physics since it provides the basic tools for the investigation of fundamental processes involving the quantum interferometry of particles. Furthermore, from a more technological perspective, the realization of a reliable source of this kind of radiation may represent a significant advance in the modern fields of quantum communication and quantum cryptography [1,2]. For this purpose the method of pump self-regularization has been adopted in the past within a few dynamical processes to provide a sub-Poissonian character of the generated light [3]. These essentially are the electron-charge-induced antibunching process acting within the excitation of a semiconductor laser and the Rabi dynamics in resonant fluorescence with excitation of single atoms in a beam, in a trap, or in a solid host [4–7]. The use of the latter process is very difficult in practice because of the delicate high-resolution spectroscopic techniques needed for the *resonant* excitation of confined single atoms in space, the hard problem of discriminating a very weak beam in the presence of a very strong one at the *same* wavelength, and, most important, the inefficiency of the process since the weak resonant scattering occurs in *all* spatial directions. In this Letter we demonstrate that these problems can indeed be overcome by a novel and efficient pump self-regularization scheme and by making use of a smart combination of optical techniques partially based on the peculiar properties of the microcavity in the context of atomic spontaneous emission (SpE) [8]. The result is a new, efficient generator of a nonclassical single-photon state that can be transformed into a quantum Fock $|n\rangle$ state generator.

Let us outline our method by referring to the single-molecule condition. A single Oxazine 720 molecule, absorbing and emitting radiation at $\lambda_p = 2\pi c/\omega_p$ and $\lambda = 2\pi c/\omega$, respectively, was excited within a virtually single-longitudinal-mode microcavity, with relevant dimension $d = m\lambda/2$, $m = 1$ (or $m =$ whole number close to 1), finesse $f = 160$, and terminated by two parallel, plane Bragg reflectors (or mirrors, $i = 1, 2$) highly reflecting at λ ($R_i \equiv |r_i|^2 \approx 1$) and transparent at $\lambda_p < \lambda$. Because of this last property, the excitation of the molecule could indeed by localized within a small volume $V = ds_p$ about equal to λ^3 at the intersection of the cavity active layer with the focal region of a 3 cm $f1$ lens collecting the excitation from a pulsed laser beam operating at λ_p. The device was excited by a collision-pulse mode-locked (CPM) laser emitting at $\lambda_p = 615$ nm a sequence of equal pulses, referred to as "δt pulses," with duration $\delta t = 0.1$ ps, energy $\epsilon = 0.12$ nJ, and rate $\nu = 1/\Delta t = 100$ MHz. The experiment was also carried out, successfully but with far more critical requirements for the parameters δt and ϵ, at $\lambda_p = 532$ nm, with a $\delta t = 5$ ns, $\nu = 20$ Hz pulsed beam SHG by a Nd-YAG Q-switched laser [9]. The selected active system was a molecular solution in ethylene glycol, a very viscous solvent at $T = 300$ K, with concentration in the range $\rho = 10^{12}$–10^{18} cm^{-3}, absorption cross section $\sigma_p(\lambda_p) = 2 \times 10^{-16}$ cm^2, and free-space SpE time $(T_1)_0 \equiv 1/\Gamma_0 \approx 4$ ns at the emission $\lambda = 702$ nm at which the microcavity was tuned. Furthermore, very important, the selected molecule had a singlet four-level optical pumping quantum efficiency η' close to 1 [9]. With a calibrated ρ and a well-stirred and highly filtered solution, to avoid any molecular clustering, the search for the single-molecule excitation condition was accomplished by transversal displacements of the lens focus in the microcavity active plane. Once found, this condition kept fairly stable in time at $T = 300$ K albeit long term stability was obtained by cooling the system at 10 K by a closed-cycle Joule-Thomson cryostat. According to a useful property of the microcavity with $m = 1$ and to its actual geometry, the light emission took place approximately over two counterpropagating plane-wave modes with vectors $\mathbf{k}$ and $\mathbf{k}' \equiv -\mathbf{k}$ orthogonal to the mirrors [9]. As far as the basic dynamics is concerned, since the quantum efficiency of the molecular absorption-emission cycle is $\eta' \approx 1$, we may say that virtually *every*

 0031-9007/96/76(6)/900(4)$06.00

pump photon extracted from the laser beam, i.e., with Poisson statistics, at λ_p is re-emitted at a *different* λ over $\mathbf{k}$ or $\mathbf{k}'$, with an antibunched character because of pump regularization, and then detected. Precisely, the overall pump regularization arises from the synergy of several processes: the *short-pulse* excitation of a *single* molecule and the "cycle self-regularization" due to the finite time taken by the excitation to cycle adiabatically through a four-level system before restoring de-excitation, as we shall see. The latter process may be somewhat related to the laser squeezing model proposed by Ritsch *et al.* [10]. The statistical character of the output beam was assessed by a Hanbury-Brown-Twiss (HBT) apparatus shown in Fig. 1 while detection was provided by two cooled (RCA31034-A) phototubes, $PM_{1,2}$, with quantum efficiencies $\xi_1 \approx \xi_2 \approx 0.12$ and average noise rate $\approx$100 Hz. The data analysis was carried out with a gated SR400 photon counter or, when necessary, by charge integration at the PM anodes. In addition to the experiment shown by Fig. 1, an equivalent Hanbury-Brown-Twiss test was also carried out by adopting an active microcavity with $R_1 = R_2$, with *no use* of any external beam splitter, the two arms of the HBT interferometer being simply provided by the two output modes $\mathbf{k}$ and $\mathbf{k}'$. These may be interpreted as corresponding to the two pure momentum states that form the basis of the quantum superposition representing any single-photon cavity excitation. This novel experimental configuration appears conceptually interesting as it suggests interpreting the microcavity as a new kind of *active beam splitter*.

Let us look more closely at the coupling process, by assuming a symmetrical cavity, $m = 1$, and a momentum-state superposition of the *polarized* emitted photon over the modes $\mathbf{k}$ and $\mathbf{k}'$ with corresponding mutually commuting operators $\hat{a} \equiv \hat{a}_{\mathbf{k}}$ and $\hat{a}' \equiv \hat{a}_{\mathbf{k}'}$ [11]. The normal-ordered Hamiltonian of the system is

$$\hat{H} = \tfrac{1}{2}\hbar\omega_p\hat{o}^\dagger\hat{o} + \tfrac{1}{2}\hbar\omega\{\hat{a}^\dagger\hat{a} + \hat{a}'^\dagger\hat{a}'\} + \tfrac{1}{2}\sum_j \hbar\omega_j\hat{\pi}_{jj} - i\hbar K_p\hat{o}^\dagger(\hat{\pi}')^- - i\hbar K'(\hat{a}^\dagger + \hat{a}'^\dagger)\hat{\pi}^- + \text{H.c.}$$

$\hat{o}$ being the single-mode pump field operator, K_p and $K' \propto \sqrt{\eta}$ appropriate coupling parameters proportional to corresponding Rabi frequencies Ω_p and Ω', η the microcavity field-enhancing factor, and $\hat{\pi}_{ij} \equiv |i\rangle\langle j|$ $(i, j = 0{-}3)$ the transition operators relative to the four-level system modeling the relevant features of the Franck-Condon dynamics of the single molecule. The system's evolution is simplified by analyzing it separately in the two 2-dimensional Hilbert subspaces spanned by states $(|1\rangle, |2\rangle), (|0\rangle, |3\rangle)$ since their respective dynamics are connected only by a rotovibrational fast relaxation process via a single coupling parameter $\gamma \equiv 1/T_2 \approx 5 \times 10^{12}\ \text{s}^{-1}$ [12]. The transition operators are $\hat{\pi}^- \equiv |0\rangle\langle 3|$ and $(\hat{\pi}')^- \equiv |1\rangle\langle 2|$ in the subspaces where the usual spin commutation relations hold for primed and unprimed operators. This allows a detailed study of the main features of the evolution of the absorption-emission cycle responsible for the self-regularization dynamics [3]. In particular, the SpE from level 3 is characterized by a cavity-enhanced, quasiexponential decay parameter $\Gamma = 2i|\Omega'|^2[\zeta(\Delta\omega) - \zeta^*(\Delta\omega)]$, where $\zeta(\Delta\omega)$ is the complex Heitler's function [13]: for our system $\gamma \gg \Gamma$. By assuming that at the initial time of any (square) δt pulse, $t = 0$, the molecular excitation is in the ground state, $\langle\hat{\pi}_{11}\rangle = 1$, the dynamics is analyzed by a Torrey-type formulation leading to the relevant statistical averages involving the field emitted and detected at the retarded time $t' = t + z/c$ by a detector placed at a distance z from the center of the cavity, on its axis $\mathbf{z}$ [14]. For instance, the intensity $\langle :\hat{I}(t'):\rangle$ radiated after excitation by a sequence of equal δt pulses, with $\delta t \ll \Gamma^{-1}$ and time interval $\Delta t \equiv v^{-1}$, $\langle\hat{\mathbf{E}}^-(z,t')\hat{\mathbf{E}}^+(z,t')\rangle = K\,\text{rep}_T\langle\hat{\pi}^+(t)\hat{\pi}^-(t)\rangle$, where $\text{rep}_T u(t) \equiv \sum_{-\infty}^{+\infty} u(t - n\Delta t)$ [15]. For $t > \delta t$ is found, to a good approximation, $\langle\hat{\pi}^+(t)\hat{\pi}^-(t)\rangle = A\{1 - B\exp(-3\gamma\delta t/2)\cos(\Omega\delta t) + C\exp(-3\gamma\delta t/2) \times \sin(\Omega\delta t)\}\exp(-\Gamma t)$, with $\Omega = [|\Omega_p|^2 - (\gamma/2)^2]^{1/2}$, $A = |\Omega_p|^2(|\Omega_p|^2 - \gamma^2)/(|\Omega_p|^2 + 2\gamma^2)^2$, $B = \frac{1}{2}(|\Omega_p|^2 - 4\gamma^2)/(|\Omega_p|^2 - \gamma^2)$, $C = -(\gamma/4\Omega)\,(7|\Omega_p|^2 - 4\gamma^2)/(|\Omega_p|^2 - \gamma^2)$, for $\Gamma \ll \gamma$. Note that with the parameters corresponding to the CPM excitation in our experiment, each laser δt pulse is a π *pulse* for the overall dynamics since $\Omega_p T_2 \approx (6\sigma_p\epsilon/\gamma\delta t s_p\hbar\omega_p)^{1/2} > 1$ and $\Omega_p\delta t \approx \pi$. Then, if a single molecule interacts with that pulse, the excitation does not have time to cycle more than once within the four-level system, leading to the emission of no more than one photon for each δt pulse. This is precisely the origin of the mechanism of self-regularization and determines the *antibunched* character of the emitted radiation [16]. With the excitation provided by longer pulses $\delta t \approx 1/\Gamma$, the π-pulse condition becomes very critically dependent on all parameters and there is the possibility of multiple cycles within δt. This would certainly lead to a Poisson-type multiple emission [9].

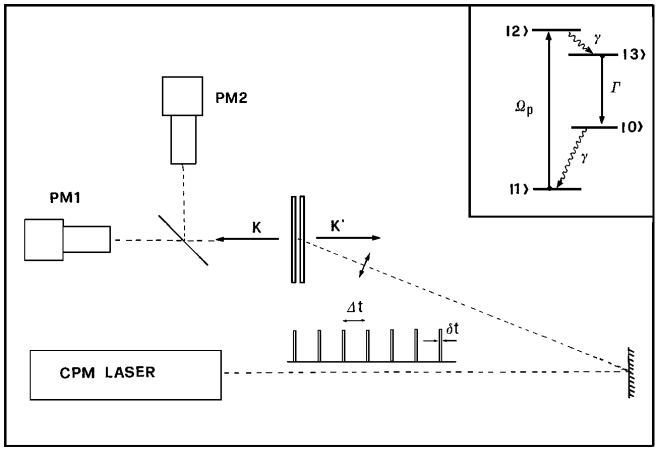

FIG. 1. Collision-pulse mode-locked laser-excited microcavity and Hanbury-Brown-Twiss apparatus.

The above analysis is completed by the evaluation of the *degree of second-order coherence*, $g^{(2)}(\tau) \equiv \langle : \hat{I}(t')\hat{I}(t'+\tau) : \rangle / [\langle : \hat{I}(t') : \rangle]^2$. This relevant quantity is evaluated by first expressing the emission intensity average $\langle \hat{\pi}^+(t+\tau)\hat{\pi}^-(t+\tau)\rangle$ as a linear superposition of molecular raising-lowering operator averages evaluated at time t. Then the second-order correlation function appearing at the numerator of $g^{(2)}(\tau)$ is evaluated with the help of the *quantum regression theorem* [16,17]. In view of the spontaneous emission dynamics involving the states $|3\rangle$, $|0\rangle$, we may write the intensity average in the simple form $\langle \hat{\pi}^+(t+\tau)\hat{\pi}^-(t+\tau)\rangle = \beta_1(\tau) + \beta_2(\tau)\langle \hat{\pi}^+(t)\hat{\pi}^-(t)\rangle$, where $\beta_1(\tau)$ and $\beta_2(\tau)$ are evaluated by solving the master equation accounting for the emission process. This leads to a straightforward evaluation of $g^{(2)}(\tau)$ which is given here for the four-level system, $t \approx 0$, $\tau < \Delta t, \delta t \ll 1/\Gamma$ in the following form: $g^{(2)}(\tau) = \overline{f}[\exp(-\Gamma\tau) - \exp(-\gamma\tau)]$, with $\overline{f} \approx 0.44\,[2\gamma\Delta t]^{-1}$ for our experimental conditions. We see that $g^{(2)}(\tau)$ is exactly equal to zero for $\tau = 0$ and very close to zero for any τ, being proportional to the small term $[2\gamma\Delta t]^{-1} \approx 10^{-4}$, as expected. In order to account formally for the experimental parameters involved in the HBT test, an equivalent quantum photodetection theory may be conveniently expressed in terms of the *coincidence parameter* α introduced by Grangier *et al.* [18]. Within the context of our work, this parameter is defined in terms of the probabilities of registering, by two detection channels 1, 2 relative to the output ports of the HBT beam splitter, coincidence and single signals for each δt pulse and within a gate interval Δt_g starting at t: $\alpha(t, \Delta t_g) \equiv \langle p_c(t, \Delta t_g)\rangle / \langle p_1(t, \Delta t_g)\rangle \langle p_2(t, \Delta t_g)\rangle$. For single-mode excitation of the beam splitter, $\Delta t_g \ll T_1$, we obtain by quantum theory [3] $\alpha(\overline{n}) \equiv \alpha(0, \Delta t_g) = \mathrm{Tr}\{\hat{\rho}\hat{N}[1 - \exp(-\xi_1\hat{a}_1^\dagger\hat{a}_1)][1 - \exp(-\xi_2\hat{a}_2^\dagger\hat{a}_2)]\}\{(\mathrm{Tr}\hat{\rho}\hat{N}[1 - \exp(-\xi_1\hat{a}_1^\dagger\hat{a}_1)])(\mathrm{Tr}\hat{\rho}\hat{N}[1 - \exp(-\xi_2\hat{a}_2^\dagger\hat{a}_2)])\}^{-1}$, where $\hat{\rho}$ represents the properties of the source field and $\hat{N}$ is the normal-ordering operator. By the n-state expansion $\hat{\rho} = \sum_n P_n|n\rangle\langle n|$, α is finally obtained for some relevant photon distributions.

(1) Chaotic:

$$P_n = \overline{n}^n/(1+\overline{n})^{1+n},$$

$$\alpha = [2 + \xi_1 T'\overline{n} + \xi_2 R'\overline{n}]/[1 + \xi_1 T'\overline{n} + \xi_2 R'\overline{n}];$$

(2) coherent:

$$P_n = \frac{\overline{n}^n}{n!}\exp(-\overline{n}), \quad \alpha = 1;$$

(3) antibunched:

$$P_n = \delta_{n,\overline{n}},$$

$$\alpha = [1 + (1 - \xi_1 T' - \xi_2 R')^{\overline{n}} - (1 - \xi_1 T')^{\overline{n}} - (1 - \xi_2 R')^{\overline{n}}]\{[1 - (1 - \xi_1 T')^{\overline{n}}][1 - (1 - \xi_2 R')^{\overline{n}}\}^{-1},$$

where $R' = |r|^2$ and $T' = |t|^2$ are the optical parameters of the (lossless) beam splitter and $\overline{n}$ the average number of photons emitted after each excitation δt pulse. By a first-order expansion of α, the second-order correlation function may be expressed in the form $g^{(2)}(0) = [\alpha - B(\overline{n})][A(\overline{n})]^{-1}$, where

$$A(\overline{n}) = (\mathrm{Tr}[\hat{\rho}\hat{a}_1^\dagger\hat{a}_1])(\mathrm{Tr}[\hat{\rho}\hat{a}_2^\dagger\hat{a}_2]) \times \{(\mathrm{Tr}\hat{\rho}\hat{N}[1 - \exp(-\xi_1\hat{a}_1^\dagger\hat{a}_1)]) \times (\mathrm{Tr}\hat{\rho}\hat{N}[1 - \exp(-\xi_2\hat{a}_2^\dagger\hat{a}_2)])\}^{-1}$$

and

$$B(\overline{n}) = \mathrm{Tr}\hat{\rho}\hat{N}\{[1 - \exp(-\xi_1\hat{a}_1^\dagger\hat{a}_1)][1 - \exp(-\xi_2\hat{a}_2^\dagger\hat{a}_2)] - \hat{a}_1^\dagger\hat{a}_1\hat{a}_2^\dagger\hat{a}_2\}\{(\mathrm{Tr}\hat{\rho}\hat{N}[1 - \exp(-\xi_1\hat{a}_1^\dagger\hat{a}_1)]) \times (\mathrm{Tr}\hat{\rho}\hat{N}[1 - \exp(-\xi_2\hat{a}_2^\dagger\hat{a}_2)])\}^{-1}.$$

According to the theory, for $n = 1$ $g^{(2)}(0) = \alpha = 0$. The parameter α is plotted in Fig. 2 for the three cases vs $\overline{n}$ and the molecular $\rho \propto \overline{n}$, for our experimental conditions. Note in Fig. 2 the good experimental verification for $\overline{n} > 1$ of the theoretical curve expressing $\alpha(\overline{n})$ in the sub-Poissonian condition, viz., implying the pure n-state distribution $P_n = \delta_{n,\overline{n}}$. These results of the HBT experiment show that an increasing sub-Poisson character of the output radiation is gradually established for ρ varying over 2 orders of magnitude, leading for $\rho \approx 7 \times 10^{14}$ cm^{-3} to the striking value $\alpha = g^{(2)}(0) = 0$ for $n = \overline{n} = 1$. This last result has been obtained at $T = 300$ K with a 50%-50% beam splitter within a run

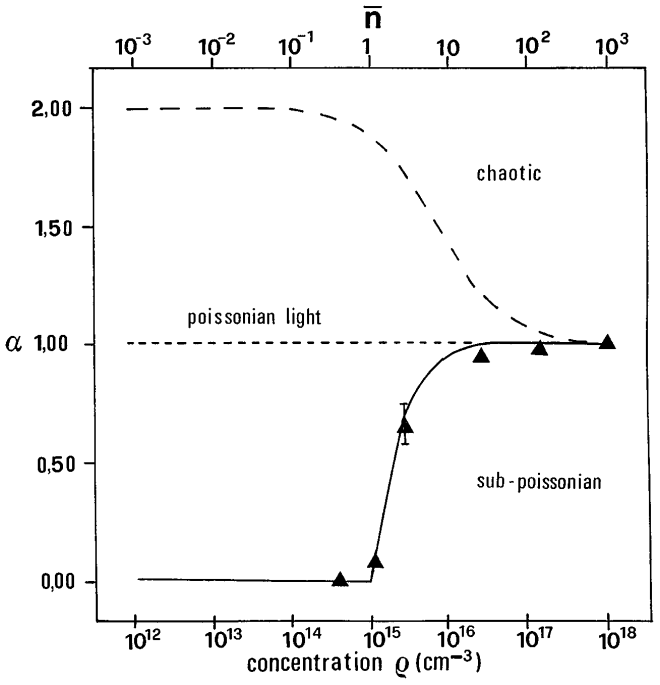

FIG. 2. Coincidence parameter $\alpha(\overline{n})$ as function of the number of photons emitted after each excitation pulse and of the molecular concentration. The time gate of the HBT apparatus is $\Delta t_g = 1$ ns and the ratio of the emission probabilities over the two output modes of the nonsymmetrical microcavity is $P(\mathbf{k})/P(\mathbf{k}') \approx 3 \times 10^2$.

involving a number of counts equal to 1.5×10^4 by each detection channel. Within this run *no coincidences* were detected. The other experimental points in Fig. 2 were determined approximately by the same number of counts.

All this provides the first demonstration that, under appropriate conditions, it is possible to conceive a macroscopic quantum device that emits, over a *single* output radiation mode, approximately a single photon per pulse, with a *quasideterministic* generation of a quantum radiation state, at a repetition rate as high as 100 MHz and with a quantum efficiency close to 1. This result leads to a still more important consequence. The single-molecule excitation process could be straightforwardly reproduced n times within the same device by multiple focusing within the macroscopic transverse extension l_t of the *same* field mode [19]. Since within that mode the SpE dynamics of the n excited molecules are strongly coupled by relativistically causal, superradiant interactions acting with a retardation time τ_t *shorter* than the coherence time τ_c of the field emitted by the microcavity with $f \gg 1$, $\tau_t = l_t/c \approx 2(\lambda/c)\sqrt{f} \ll \tau_c \approx (\lambda/c)f$, then the emitted, indistinguishable n single photons do belong to the *same* space-time extension of the output field mode, i.e., they form a quantum Fock $|n\rangle$ state [20]. The experimental realization of this condition would certainly determine a new exciting endeavor within the quantum optics community. The preliminary results of our investigation in this direction are quite encouraging.

We acknowledge useful discussions with P. W. Milonni, J. D. Franson, and Y. Shih.

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