Bose-Einstein Partition Statistics in Superradiant Spontaneous Emission

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We report the realization of the *spatial* counterpart of the Dicke superradiance. The new process is revealed by the realization of the spatial quantum partition statistics within the detection of photons emitted in sub-Poissonian regime by an active microcavity excited by ultrashort pulses. The superradiant enhancement of the *time* decay of the dipole excitation has also been investigated.

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Radiation from a source composed of two atoms may exhibit very interesting features. If the two atoms are situated in free space at a mutual distance $R \leq \lambda$, the rate of spontaneous emission (SE) at the wavelength (wl) λ can be as high as the double of the corresponding rate of a single atom. This is the most relevant aspect of the "superradiance" *time* QED process [1,2]. The present Letter reports on a new approach to superradiance by adoption of a new technique, i.e., by trapping the atoms within a biplanar high-finesse microcavity, a device that has recently been found to behave as a source of nonclassical radiation [3]. Here this method leads to the demonstration of the *spatial* counterpart of the superradiance process by the discovery of an unexpected quantum statistical distribution of the photons emitted by the microcavity *as a whole,* over the two allowed external modes **k** and **k**$'$. By this process the active microcavity, in spite of being a macroscopic device, reveals its genuine nature of quantum nanostructure ascribable to its peculiar particle-confining topology more than to the intrinsic quantum nature of the confined particles themselves. To our knowledge, a spatial collective phenomenon of this kind has not been investigated before, either theoretically or experimentally.

Two independent femtosecond pulses with wavelength $\lambda_p = 615$ nm and duration $\delta t = 80$ fs were focused by a common lens within two transverse focal spots with diameter $\varphi = 10$ μm on the symmetry plane $\mathbf{Z} = 0$ of a planar active microcavity with relevant dimension $d = \frac{1}{2}\lambda$ to excite two dipoles located at an externally adjustable mutual transverse distance **R** along the spatial **Y** axis (cf. Fig. 1). The wl of the SE radiation was $\lambda = 700$ nm with an emission bandwidth $\Delta\lambda = 0.2$ nm, due to the spectral cavity filtering. The microcavity was terminated by two equal mirrors with reflectivity $\mathcal{R} \equiv |r|^2 = 0.9990$ at wl λ and the cavity "finesse," $f = 3000$, determined the "coherence time" of the emitted particles, $\tau_c \approx 1$ ps. The active medium filling the cavity consisted of a $10^{-5} M$/liter concentration of Oxazine 725 molecules in a polymethyl methacrylate solid film, cooled at liquid nitrogen temperature. An important dynamical feature of the active microcavity consists of the establishment of an interdipole coupling over a *macroscopic* "transverse coherence length" $\ell_c = 2\lambda\sqrt{fm}$ which corresponds to the effective radius of the Gaussian-like electromagnetic emission mode in the cavity [4,5]. In a recent experiment we found that the dynamics of two excited dipoles placed at a distance $R \equiv |\mathbf{R}| \leq \ell_c$ is indeed causally connected within a retardation time $\tau \lesssim \ell_c/c$ [6]. In the present work the regime of superradiant photon emission has been investigated by adopting two different experimental interferometric configurations: Fig. 1. Namely, configuration A (CA) in which the

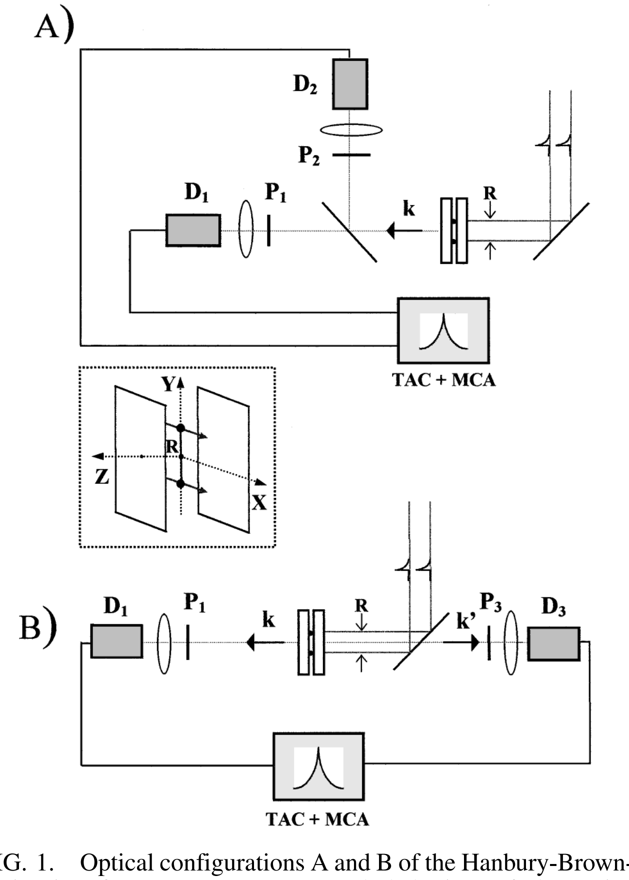

FIG. 1. Optical configurations A and B of the Hanbury-Brown-Twiss interferometers. Inset: Exploded view of the active microcavity.

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measurement apparatus consisted of the detectors D_1 and D_2 coupled via an external 50/50 beam splitter to only one output mode $\mathbf{k}$ of the microcavity, and configuration B (CB) in which the apparatus consisted of D_1 and D_3 coupled to both output modes $\mathbf{k}$ and $\mathbf{k}'$. When a *single* active spot was excited in the cavity, the *single photon* condition was tested carefully by appropriate Hanbury-Brown-Twiss tests carried out by either CA or CB. The tests showed a striking evidence of the nonclassical antibunching process within the emission process [3,7]. As an example, by adopting CA we found a value of the degree of second-order coherence $g^{(2)} = 4.4 \times 10^{-2}$ on the basis of the following results: number of detected D_1-D_2 coincidences = 3, detected "singles" = 3810 and 3250, and statistical sample = 1.8×10^5 laser excitations. There the photons were detected within time windows of 5 ns following each excitation. The detectors consisted of single photon-counting modules EGG-SPCM200 with quantum efficiencies ~65%.

Moving to the main experiment involving *couples* of photons, the time measurements were carried out by a time-to-amplitude converter (TAC) with the detectors D_1 and D_2 (or D_3) acting as start/stop devices, and by a multichannel analyzer (MCA). Because of the random orientation of the active molecules, the output radiation was found slightly (20%) polarized along the linear polarization of the excitation beams [8]. The emission properties of the active dipoles along the orthogonal directions $\mathbf{X}$ and $\mathbf{Y}$ were investigated by filtering the output radiation with adjustable polarization analyzers P_j. The polarization of the excitation pulses was set oriented along $\mathbf{X}$. Both CA and CB were adopted: by CA we investigated the photon emission over the single output mode $\mathbf{k}$, while by CB we investigated the emission over both modes $\mathbf{k}$ and $\mathbf{k}'$. The first experiment, carried out by CA, consisted of the measurement of the temporal evolution of the normalized second-order field correlation function: $F(\tau) \propto \langle \hat{\mathbf{E}}^-(t)\hat{\mathbf{E}}^-(t+\tau)\hat{\mathbf{E}}^+(t+\tau)\hat{\mathbf{E}}^+(t)\rangle$ being τ the time delay between two photodetections. Precisely, we measured the *normalized* quantity $F(\tau) = \lim_{T\to\infty} g^{(2)}(\tau) \times [T^{-1}\int_0^T g^{(2)}(\tau)\, d\tau]^{-1}$. As we shall see later, by assuming the exponential decay for the SE probability, $F(\tau)$ may be interpreted as the normalized time probability distribution of detecting $n_{\mathbf{k}} = 2$ photons emitted over the output mode $\mathbf{k}$ and $n_{\mathbf{k}'} = 0$ photons over the mode $\mathbf{k}'$. Because of the normalization of $F(\tau)$ we may write $F(0) = A \times P(2,0)$, where this last quantity is the probability of *simultaneous emission* of two photons over the output mode $\mathbf{k}$ and zero photons over $\mathbf{k}'$. A is a proportionality coefficient. A similar argument can be applied when $F(\tau)$ is determined by CB. In this case we get $F(0) = A \times P(1,1)$. The inset of Fig. 2 shows two normalized MCA curves obtained for two values of the interdipole spacing, $R = 0.33\ell_c = 25\ \mu$m and $R = 7.2\ell_c = 570\ \mu$m, being $\ell_c = 77\ \mu$m. In Fig. 2 we have reproduced the same results in a semilog scale for small values of τ, by adding a further set of data taken with $R = 2.9\ell_c = 230\ \mu$m. These results were obtained with both polarizers P_j oriented along the $\mathbf{X}$ axis. The experimental results show an enhancement $\eta \approx 1.8$, of the SE rate for two interacting dipoles placed at a mutual distance $R \lesssim \ell_c$. When P_1 and P_2 were set mutually orthogonal, the enhancement effect disappeared and the SE rate coincided with that of a single cavity-confined dipole [9]. When both analyzers P_j were oriented along the $\mathbf{Y}$ axis, corresponding to the less efficient head-on dipole-dipole interaction, the SE enhancement effect was reduced to $\eta \approx 1.2$.

In order to get a better insight into the process, let us study the time evolution of the field radiated by two dipoles, parallel to the $\mathbf{X}$ axis, excited at the time $t_0 = 0$ and observed at a later time t by a detector located on the $\mathbf{Z}$ axis at a distance $Z \gg \lambda$ from the center of a lossless microcavity. In the Heisenberg representation, the field can be expressed in terms of the dipole transition operators $\hat{\pi}_A(t)$ and $\hat{\pi}_B(t)$ [7,9]:

$$\hat{\mathbf{E}}^+(Z,t) = -\Theta(1+r)(1-|r|^2)^{1/2} \times \left[\hat{\pi}_A\left(t - \frac{Z}{c}\right) + \hat{\pi}_B\left(t - \frac{Z}{c}\right)\right]\sum_{n=0}^{\infty} r^{2n}. \quad (1)$$

Here the effect of multiple intracavity reflections is considered, τ is the reflection coefficient of the mirrors at normal incidence, and Θ is a constant. We may insert the above expression into the definition for $F(\tau)$ and make use of the ansatz $\hat{\pi}(t) = \hat{\pi}(0)\exp\{-[i\frac{2\pi c}{\lambda} + \frac{1}{2}\Gamma(R)]t\}$, implying that no causal interdipole interaction is established at $t_0 = 0$ [5]. By accounting for the antibunching character of the output radiation, the second-order correlation function may be written as

$$F(\tau) \propto \sum_{i\neq j}[\langle\hat{\pi}_i^\dagger(t)\hat{\pi}_j^\dagger(t+\tau)\hat{\pi}_j(t+\tau)\hat{\pi}_i(t)\rangle + \langle\hat{\pi}_i^\dagger(t)\hat{\pi}_j^\dagger(t+\tau)\hat{\pi}_i(t+\tau)\hat{\pi}_j(t)\rangle],$$

for $i,\ j = A, B$. By replacing in the sums the ensemble averages with time averages, we find $F(\tau) \propto \exp[-\Gamma(R)|\tau|]$. The following explicit expression of $\Gamma(R)$, for the case of two dipoles oriented along $\mathbf{X}$ and expressed as a function of the *free-space* SE rate $\Gamma \equiv (T_{\mathrm{SE}}^{-1})$, is found [5]:

$$\Gamma(R) = \Gamma\left\{1 + \frac{3}{2k^3}\left[\sin(kR)\left(-\frac{1}{R^3} + \frac{k^2}{R}\right) + \cos(kR)\frac{k}{R^2}\right]\theta(ct - R) + \frac{3}{k^3}\sum_{n=1}^{\infty}(-|r|)^n \times \left\{\left[\sin(knd)\left(-\frac{1}{(nd)^3} + \frac{k^2}{nd}\right) + \cos(knd)\frac{k}{(nd)^2}\right]\theta(ct - nd) + \left[\sin(kR_n)\left(-\frac{1}{R_n^3} + \frac{k^2}{R_n}\right) + \cos(kR_n)\frac{k}{R_n^2}\right]\theta(ct - R_n)\right\}\right\}, \tag{2}$$

where $R_n = \sqrt{R^2 + (nd)^2}$ and $\theta(ct - x)$ are Heaviside unit step functions accounting for relativistic causality in the interdipole interactions. It is found that for maximum superradiance ($kR \sim 0$) the value of $\Gamma(R)$ is *twice* as large as the value Γ_∞ of the case of two independent dipoles ($R \gg \ell_c$). The dashed lines shown in Fig. 2 are drawn in the semilog scale according to Eq. (2). The experimental results are in agreement with theory.

The above results show that the peculiar topology of the microcavity is instrumental in the determination of the *time* behavior of a SE decay process within an interdipole interaction. Indeed, the *mesoscopic* character of the microcavity is ascribable to the fact that the de Broglie wavelength λ of the confined particle, the photon, is of the order of the relevant dimension d of the confining device. This is a common feature of all nanostructures that exhibit quantum properties. In this perspective we thought that the *spatial* behavior of some relevant dynamical process should also be affected by the peculiar particle-confining properties of the device. In the new experiment, we investigated the spatial statistical distribution of the photon pairs emitted over the two output modes $\mathbf{k}$ and $\mathbf{k}'$.

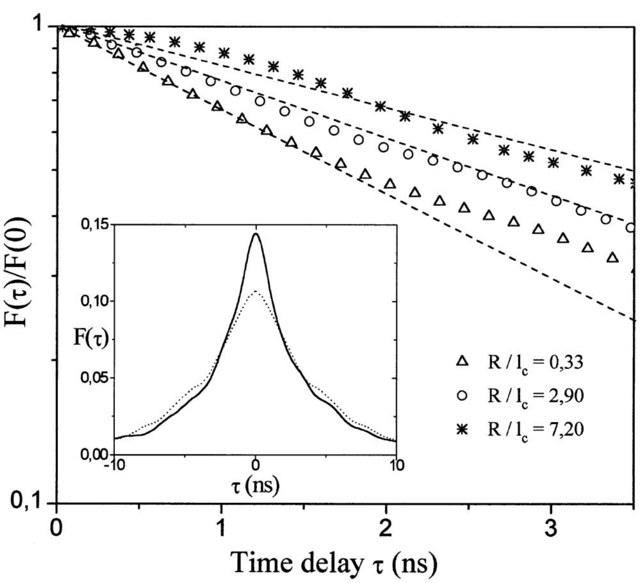

FIG. 2. Semilog plot of the two photon normalized correlation function $F(\tau)/F(0)$ as a function of the delay τ between the detected photons for $R/\ell_c = 0.33$ (Δ), 2.9 ($\circ$), and 7.2 ($*$). Inset: Experimental normalized distributions $F(\tau)$ for $R/\ell_c = 0.33$ (solid curve) and $R/\ell_c = 7.2$ (dotted curve).

The experiment was carried out by adopting both CA and CB and by measuring the probabilities $P(2,0)$ and $P(1,1)$ of the simultaneous photodetections realized by the couples $D_1 - D_2$ and $D_1 - D_3$, respectively. By assuming a "classical," Maxwell Boltzmann, partition statistics we should expect that the probability of single photon detection on both modes is *twice as large* as the one of two photon detection on the same mode by a single detector: $P(1,1) = 2P(2,0)$. The experimental results given in Fig. 33 show that this is indeed verified for $R \gg \ell_c$. However, by carrying out the experiment with $R \lesssim \ell_c$, the relative values of the probabilities are found to converge toward the common value $P(1,1) = P(2,0)$. We shall see shortly that this implies that the quantum Bose-Einstein (BE) partition process determines the overall photoemission from the microcavity, a device that then behaves like a *two photon* "quantum lamp."

We may explain this remarkable spatial quantum phenomenon as follows. The two equally polarized photons are emitted over a *single* internal *standing-wave* (sw) $\mathbf{k}$ mode of the microcavity if the condition $R \ll \ell_c$ is satisfied. It consists of the linear superposition of the two traveling wave (tw) modes associated with the *internal* momenta $\mathbf{p} = \hbar\mathbf{k}$, $\mathbf{p}' = \hbar\mathbf{k}' = -\mathbf{p}$. The field operators acting on these tw modes are identified by the labels ℓ

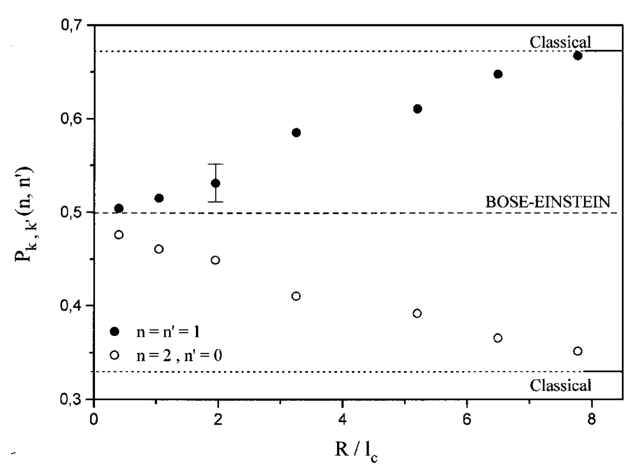

FIG. 3. Two photon partition probabilities $P(1,1)$ and $P(2,0)$ vs R/ℓ_c. The values of the probabilities are referred to the total number of detection events actually registered in the experiment. By reporting the data obtained by configuration A we have accounted for the effect of the beam splitter.

or r, respectively. The density operator representing the state of the field in the condition $R \ll \ell_c$ is then given by $\rho_1 = \pi^{-2} \iint d\varphi\, d\varphi' \cos^2\varphi \cos^2\varphi' \times (\hat{a}_\ell^\dagger + e^{i\varphi}\hat{a}_r^\dagger) \times (\hat{a}_\ell^\dagger + e^{i\varphi}\hat{a}_r^\dagger)\,|\text{vac}\rangle\langle\text{vac}|\,(\hat{a}_\ell + e^{-i\varphi'}\hat{a}_r)\,(\hat{a}_\ell + e^{-i\varphi}\hat{a}_r)$. There the vacuum field is $|\text{vac}\rangle \equiv |0,0\rangle \equiv |\ell = 0, r = 0\rangle$, the phases $\varphi = \pi Z/d$, $\varphi' = \pi Z'/d$ account for the random position of the emitting dipoles along the longitudinal coordinate Z, and the distribution function $\pi^{-1}\cos^2\varphi$ expresses the SE enhancement in the plane microcavity with $d = \frac{\lambda}{2}$ as a function of Z [9]. After integration, and normalization is found, $\rho_1 = \frac{1}{3}(|2,0\rangle \times \langle 2,0| + |1,1\rangle\langle 1,1| + |0,2\rangle\langle 0,2|)$, where, e.g., $|2,0\rangle \equiv |\ell = 2, r = 0\rangle$. For a larger transverse distance between the emitting dipoles, $R \gg \ell_c$, the emission takes place over two *distinct*, cavity sw modes, i.e., over two couples of tw modes ℓ, ℓ' and r, r'. The system is now represented by $\rho_2 = \pi^{-2} \iint d\varphi\, d\varphi' \cos^2\varphi \cos^2\varphi' (\hat{a}_\ell^\dagger + e^{i\varphi}\hat{a}_r^\dagger)\,(\hat{a}_\ell'^\dagger + e^{i\varphi'}\hat{a}_r'^\dagger)\,|\text{vac}\rangle\langle\text{vac}|\,(\hat{a}_\ell' + e^{-i\varphi'}\hat{a}_r')\,(\hat{a}_\ell + e^{-i\varphi}\hat{a}_r)$ where $\hat{a}_\ell$, $\hat{a}_\ell'$, $\hat{a}_r$, $\hat{a}_r'$ are acting over the corresponding modes ℓ, ℓ', r, r', and the vacuum field is $|\text{vac}\rangle \equiv |0,0;0,0\rangle \equiv |\ell = 0, \ell' = 0; r = 0, r' = 0\rangle$. In this case is found $\rho_2 = \frac{1}{4}(|1,1;0,0\rangle\langle 1,1;0,0| + |1,0;0,1\rangle\langle 1,0;0,1| + |0,1;1,0\rangle\langle 0,1;1,0| + |0,0;1,1\rangle \times \langle 0,0;1,1|)$. This analysis leads immediately to the predictions of the experiment. If the *single* cavity sw-mode condition is realized, $R \ll \ell_c$, we obtain $P(2,0) = \langle 2,0|\rho_1|2,0\rangle = \frac{1}{3}$, $P(0,2) = \langle 0,2|\rho_1|0,2\rangle = \frac{1}{3}$, $P(1,1) = \langle 1,1|\rho_1|1,1\rangle = \frac{1}{3}$. This nonclassical result reproduces the BE statistics according to which the probability of distributing N *indistinguishable* particles among G "boxes" is *independent* of the set of occupancies of the boxes, here indicated by $\{n_i\}$, and is given by $P\{n_i\} = [(G - 1)!\,N!/(G + N - 1)!]$ [10]. On the other hand, if the distance between the dipoles is large, $R \gg \ell_c$, the two photons are emitted over two *distinct* cavity sw modes and as such they become *distinguishable*. In this case the following probabilities are found: $P(2,0) = \langle 1,1;0,0|\rho_2|1,1;0,0\rangle = \frac{1}{4}$, $P(0,2) = \langle 0,0;1,1|\rho_2|0,0;1,1\rangle = \frac{1}{4}$, $P(1,1) = \langle 0,1;1,0|\rho_2 \times |0,1;1,0\rangle + \langle 1,0;0,1|\rho_2|1,0;0,1\rangle = \frac{1}{2}$. As expected, this result is in agreement with the classical Maxwell-Boltzmann statistics: $P\{n_i\} = G^{-N}N!/(n_l!\,n_r!)$, where the boxes, labeled by $i = l, r$ express detection on either side of the cavity, as said. Note that the two statistical formulas just given reproduce exactly the results of the experiment for $G = 2$, $N = n_l + n_r = 2$.

In summary, we have found that, for $R/\ell_c \ll 1$, the photons tend to be emitted both at the same time and over the *same spatial* mode of the active cavity. This represents a relevant conceptual contribution to the well established picture of the *superradiance* process [1]. Furthermore, we have given an insightful picture at a fundamental microscopic level of the very first stages of the collective dynamics of the *thresholdless microlaser* [11] and of the vertical cavity surface emitting laser [12].

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