BOSE - EINSTEIN PARTITION
STATISTICS OF PHOTONS EMITTED
FROM A SUPERRADIANT ACTIVE
MICROCAVITY

E. De Angelis, F. De Martini, and P. Mataloni
Dipartimento di Fisica and
Istituto Nazionale di Fisica della Materia
Università di Roma ”La Sapienza”, Roma, 00185 - Italy

Abstract

We report the results of the first investigation on the superradiant temporal and spatial quantum dynamics of two dipoles excited in a planar symmetrical microcavity by a controlled femtosecond two-pulse excitation. A superradiant enhancement of the time decay of the dipole excitation for a decreasing inter-dipole transverse distance $R$ has been found. Furthermore, the photon partition statistics of the emitted field is found to exhibit a striking quantum behaviour for $R \leq l_c$, the transverse extension of the single allowed microcavity mode.

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

It has been demonstrated that the planar active microcavity behaves as a reliable source of nonclassical radiation when small ensembles of fluorescent molecules are isolated within the optical resonator and excited by a sequence of femtosecond laser pulses [1, 2]. In these conditions the ultrashort pulse excitation forbids the dynamical recycling between the lower and the excited molecular levels leading to a strictly single photon emission over one of the two allowed external output modes, $\mathbf{k}$ and $\mathbf{k}'$, of the microcavity with relevant ”microscopic” dimension $d = m \frac{\lambda}{2}$ and $m = 1$. We can double this process by focusing two independent femtosecond pulses, within a transverse
focal spot dimension $\sim \lambda_p$, the excitation wavelength (wl), to excite two dipoles located within the microcavity at a mutual transverse distance $R$, on the symmetry plane $Z = 0$. The problem of the transverse quantum interaction between two dipoles in a cavity, generally with different spatial orientations, has been so far only theoretically addressed in terms of superradiance in atomic spontaneous emission (SE) [3, 4]. In recent work we have found that a typical superradiant regime is established in the microcavity when $R \equiv |R|$ is shorter than $\ell_c = 2\lambda\sqrt{fm}$, the ”transverse coherence length”, i.e., the effective radius of the gaussian - like e.m. mode in the cavity, being $\lambda$ the emitted wavelength and $f \gg 1$ the cavity ”finess” [5]. In particular we found experimentally that the dynamics of the two dipoles within the microcavity is causally connected within a retardation time $\tau \lesssim \ell_c/c$ which is in the sub-picosecond time i.e., corresponding precisely to the time resolution allowed by a standard femtosecond laser technique [6, 7]. It is worth noting in this connection that in free-space, i.e., in absence of cavity confinement, the corresponding two-atom superradiant coupling can be established only over a ”microscopic” inter-atomic transverse distance $R \approx \lambda$. This makes impossible the selective, independent optical excitation of the interacting fixed dipoles as well as any controlled photon emission over a limited number of modes [3, 4]. Furthermore, the peculiar, highly favorable topological properties of the planar microcavity in superradiance investigations are further enlightened by considering that the adoption of the spherical or confocal resonators, of general use in modern cavity QED, again is associated with a ”microscopic” size of $\ell_c$ thus reproducing closely the free-space impossibility condition. In our present experiment two laser beams were focused by a common lens with f.l. 20 cm on the active plane of a single mode symmetrical microcavity in two focal spots with diameter $\varphi = 10 \mu m$ at an externally adjustable mutual transverse distance $R$ along the spatial $Y$ axis, (cfr: Figure 1). The corresponding two excitation optical pulses, generated by an amplified colliding pulse mode-locking (CPM) dye laser withwl $\lambda_p = 615 nm$ and duration $\delta t = 80 fs$. The active medium consisted of a $10^{-5}M/\ell$ concentration of Oxazine 725 molecules in a matrix of polymethyl methacrylate (PMMA) solid film, cooled at liquid nitrogen temperature. The wl of the emitted radiation was $\lambda = 700 nm$. The microcavity consisted of a single-longitudinal mode Fabry-Perot interferometer, terminated by two parallel, plane multilayer dielectric equal mirrors, highly reflecting ($R \equiv |r|^2 = .9990$) at $\lambda$ and highly transparent ($T = .98$) at $\lambda_p$. The cavity ”finess” was $f = 3000$. This value determines the microcavity storage time which is equal
to the "coherence time" of the emitted particles: \( \tau_c \approx 1 \text{ ps} \). The condition of single-photon emission from the microcavity following any single laser pulse excitation, indeed a critical condition in the context of the present work, was carefully tested experimentally by use of suitable Hanbury Brown-Twiss (HBT) interferometric configurations involving each or, alternatively, both output modes, \( k \) and \( k' \). The HBT coincidence rate, evaluated as the ratio between the number of spurious two-detector coincidences and the number of detected "singles" per second, was found less than \( 10^{-3} \). The output photons were detected by cooled, avalanche single photon-counting modules EGG-SPCM200. They are indicated by \( D_1, D_2, D_3 \) in Figure 1. The typical quantum efficiency of the three equal detectors was 70%. The significant time measurements were carried out by feeding a time-to-amplitude converter (TAC) with the standard TTL output pulses of couples of detectors. The TAC (Silena 7412) was connected to a Multichannel Analyzer (MCA) (Silena 7923-2048). The output radiation, spectrally filtered by the microcavity within a bandwidth \( \Delta \lambda = 0.2 \text{ nm} \), was focused into the active surface of the detectors, with diameter = 100 \( \mu \text{m} \), by 5 cm f.l. lenses. Because of the random orientation of the active molecules the output radiation was found slightly (20%) linearly polarized along the (linear) polarization of the excitation laser beams. In order to investigate the emission properties of the active dipoles along the orthogonal transverse spatial directions \( X \) and \( Y \), the output radiation detected by each \( D_j \) was filtered by adjustable optical polarization analyzers \( P_j \). The polarization of the excitation pulses was set oriented along \( X \). Two different experimental configurations, involving two laser pulse excitation, were investigated. Configuration A) in Figure 1: by adoption of \( D_1 \) and \( D_2 \) as start and stop devices for the TAC, we could measure photon pairs emitted over the single output mode \( k \), i.e. on one side of the microcavity. Configuration B): the adoption of \( D_1 \) and \( D_3 \) as start-stop devices, allowed direct HBT investigations on both output modes \( k \) and \( k' \) of the microcavity, here used as a kind of active beam splitter. The first experiment, involving the configuration A) in Figure 1, consisted of the measurement of the temporal evolution of the second-order electric field correlation function \( F(\tau) = \langle \hat{E}^- (t) \hat{E}^- (t+\tau) \hat{E}^+ (t+\tau) \hat{E}^+ (t) \rangle > \propto P_{k,k'}(2,0) \), where \( \tau \) represents the time difference between the output pulses released by the two detectors and \( P_{k,k'}(2,0) \) is the probability of detection of 2 photons over the mode \( k \) and zero photons over the mode \( k' \). The inset of Figure 2 shows two typical, normalized MCA curves obtained for two extreme values of the inter-dipole spacing, \( R \). The solid curve corresponds to \( R = 0.33 \ell_c = 25 \)
µm, being $\ell_c = 77 \mu m$ for our experiment. The dotted curve corresponds to $R = 7.2 \ell_c = 570 \mu m$. In Figure 2 we have reproduced the same results in a semilog scale, for small values of $\tau$, by adding a further intermediate set of data, taken for $R = 2.9 \ell_c = 230 \mu m$. All these results have been obtained with both polarizers $P_j$ oriented along a common spatial direction $X$. The experimental results show a marked enhancement, of a factor 1.8, of the rate of spontaneous emission (SE) by two interacting dipoles which are selected to be mutually parallel and placed at a mutual distance $R \lesssim \ell_c$. The enhancement effect has been found to disappear when $P_1$ and $P_2$ are set mutually orthogonal, i.e., along the directions $X$ and $Y$. In this case the rate of spontaneous emission is found equal to the SE value $\Gamma_\infty \equiv T^{-1}$, of a single atom in the microcavity [9] and to the lowest value obtained in the previous case, i.e., for $R >> \ell_c$. In addition, when both analyzers $P_j$ are oriented along the $Y$ axis, corresponding to the less efficient head-on dipole-dipole interaction, the SE enhancement effect has been found to be reduced of about a factor 1.2. In summary, all these results realize the expected features of a superradiant model involving two dipoles e.m. interacting in an optical microcavity.

In order to get a better insight into this process, let us consider here the simple case of the time evolution of the field radiated by two equal dipoles, both parallel to the $X$ axis, excited instantaneously at the time $t_0 = 0$ and observed at a later time $t$ by a detector located on the $Z$ axis at a distance $Z >> \lambda$ from the center of a symmetrical, lossless microcavity. In the Heisenberg representation, the field can simply be expressed in terms of the transition operators $\hat{\pi}_A(t)$ and $\hat{\pi}_B(t)$ of the two dipoles $A$ and $B$ [9, 10]:

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

where the effect of multiple intracavity reflections is considered, $r$ is the value of reflection coefficient of the mirrors at normal incidence and $\Theta$ is a constant. We may insert the above expression into the definition for $F(\tau)$, by assuming that the atomic operators labelled by $A$ and $B$ mutually commute at first order and by taking into account only the terms in which each $\hat{\pi}$ operator is multiplied by a $\hat{\pi}^\dagger$ corresponding to either atom $A$ or $B$ [10]. We may further make use of the ansatz $\hat{\pi}(t) = \hat{\pi}(0) \exp \left[ -(i \frac{2\pi c}{\lambda} + \frac{1}{2} \Gamma(R)) t \right]$ implying that no causal inter-dipole interaction is established at $t_0 = 0$ [3, 4].
At last, the second order correlation function may be written as: $F(\tau) \propto \sum_i \langle \hat{\pi}_i^\dagger(t)\hat{\pi}_i^\dagger(t+\tau)\hat{\pi}_i(t+\tau)\hat{\pi}_i(t) \rangle + \sum_{i\neq j} \left[ \langle \hat{\pi}_i^\dagger(t)\hat{\pi}_j^\dagger(t+\tau)\hat{\pi}_j(t+\tau)\hat{\pi}_i(t) \rangle \right. \\
+ \left. \langle \hat{\pi}_j^\dagger(t)\hat{\pi}_i^\dagger(t+\tau)\hat{\pi}_i(t+\tau)\hat{\pi}_j(t) \rangle \right], \text{ for } i, j = A, B. \text{ The first sum vanishes}
\text{ owing to the antibunched character of the emitted radiation. By replacing}
\text{ in the second sum the ensemble averages with time averages, the expected}
\text{ result is found by a simple integration: } F(\tau) \propto \exp \left( -\Gamma(\tau) |\tau| \right). \text{ This result}
\text{ may be compared with the experimental MCA output data reported in the}
\text{ inset of Figure 2. The explicit expression of } \Gamma(R), \text{ evaluated by a fully rela-
\text{ativistic quantum field theoretical analysis, is expressed as a function of the}
\text{ free-space SE rate } \gamma = \frac{1}{2}(T_{SE}^{-1}) \text{ of a single dipole [3]. In the case of coupled}
\text{ dipoles commonly oriented along } X \text{ the following result is found:}

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

where $R_n = \sqrt{R^2 + (nd)^2}$ and $\theta(cx - x)$ are Heaviside step functions accounting for relativistic causality in the establishment of the inter dipole interactions. By adopting the actual values of the microcavity parameters within an explicit numerical evaluation, it is found that the value of $\Gamma(R)$ corresponding to the condition of maximum superradiance $(R = 0)$ is twice as large as the value of the case of two independent dipoles $(R >> \ell_c)$. This is in good agreement with the experimental results for SE time decay, as shown by Figure 2.

The above results show once again that the peculiar topology of the microcavity is instrumental in the determination of the time behavior of a quantum SE decay process within an inter-atomic interaction. It is easy to recognize that the mesoscopic character of the device is precisely ascribable to the fact that the De Broglie wavelength $\lambda$ of the confined particle, the photon, is of the order of the relevant dimension $d$ is of the confining device. This is a
common characteristics of all nanostructures that exhibit quantum properties. In this perspective, it is expected that also the *spatial* behaviour of some relevant dynamical process should be affected by the peculiar quantum properties of the device.

We may look for instance at the spatial statistical distribution of the couples of photons emitted over the two allowed microcavity output modes \( k \) and \( k' \) under corresponding couples of excitation laser pulses. This process has been investigated with the same microcavity by both experimental configurations A) and B), Figure 1, and for very small time delay \( \tau \approx 0 \). Precisely, we have measured the probability \( P_{k,k'}(2,0) \) of the simultaneous photodetections realized by \( D_1 \) and \( D_2 \) coupled to one external output mode \( k \), and the probability \( P_{k,k'}(1,1) \) of the simultaneous photodetections realized by \( D_1 \) and \( D_3 \) coupled to the counterpropagating external output modes \( (k, k') \).

By assuming a "classical" Maxwell-Boltzmann partition statistics, and by accounting for the couples of detection events, we expect: \( P_{k,k'}(2,0) = \frac{1}{3}, \) \( P_{k,k'}(1,1) = \frac{2}{3} \). This implies that the value of \( \Gamma(R) \) measured by the experimental configuration B) is twice as large as the one measured by configuration A). The experimental results given in Figure 3 show that this is indeed verified for a large inter-dipole distance: \( R >> \ell_c \). However, these results also show that, for shorter distances \( R \lesssim \ell_c \), the relative values of the probabilities converge toward the common value: \( P_{k,k'}(1,1) = P_{k,k'}(2,0) = \frac{1}{2} \). This implies that a quantum Bose-Einstein (BE) partition process determines the photoemission over the external modes of the microcavity, a device that is then found to behave like a two photon "quantum lamp". The realization of a quantum statistical photon distribution law at the output of an optical cavity has never been investigated before. We may try to explain this remarkable quantum phenomenon by the following simplified argument. Because of the condition \( R \ll \ell_c \), the two photons are emitted over the allowed stationary mode of the microcavity, which consists of the superposition of the two travelling - wave modes associated with the *internal* momenta \( h k \) and \( h k' = -h k \). Since the cavity mode identifies the spatial extent of the minimum - uncertainty application of the Heisenberg principle of the tridimensional photon dynamics, the two particles are in principle dynamically "indistinguishable" and then the *internal* two photon state is expressed in terms of the momentum eigenvectors by: \( |\Psi\rangle_{in} = 3^{-\frac{1}{2}}(|2,0\rangle + |1,1\rangle + |0,2\rangle \),

where: \( |x,y\rangle \equiv |x\rangle_k |y\rangle_{k'} \). Note that, according the BE partition law, the state \( |1,1\rangle \) is counted only once within the structure of \( |\Psi\rangle_{in} \). Consider now that the two photon transmission throught the mirrors of the lossless
microcavity formally corresponds to a simple unitary transformation leading to: $|\Psi_{\text{out}}\rangle \propto |\Psi_{\text{in}}\rangle$. In the expression of $|\Psi_{\text{out}}\rangle$, the function accounting for the two photon state outside the cavity, $\hbar k$ and $\hbar k' = -\hbar k$ are to be interpreted as external momenta, i.e. affecting the particle dynamics outside the cavity. This leads to the quantum result found experimentally for $R \ll \ell_c$: $P_{k,k'}(1,1) \equiv |\langle 1,1 | \Psi_{\text{out}} \rangle|^2 = P_{k,k'}(2,0) \equiv |\langle 2,0 | \Psi_{\text{out}} \rangle|^2 = 1/2$. On the other hand, the couple of photons created by two distant dipoles, $R \gg \ell_c$ do not belong to the same gaussian-like internal stationary microcavity mode and then the above indistinguishability condition is lost. As this condition is reproduced outside the cavity within the detection process it leads eventually to the realization of the classical result $P_{k,k'}(1,1) = 2P_{k,k'}(2,0)$. In summary we have found that, for $R/\ell_c \ll 1$, the two photons tend to be emitted at the same time and over the same spatial output mode of the microcavity. This striking spatio-temporal ”photon dragging” process, found here under a strictly controlled simultaneous two-photon emission, could also be detected by exciting a large unknown number of active molecules in the single mode microcavity. In this case the quantum character of the multiphoton statistical process can be identified by the experimental determination of the two-channel ”quantum noise function” introduced in a different context by work \[1\]. A more detailed description of the experiment and a more extended theoretical analysis will be given in a following paper. This research was carried out under the CEE-TMR Contract ERBFMRXCT96-0066. We also thank MURST and INFM (Contract No PRA97-cat) for funding.

References

[1] F. De Martini, G. Di Giuseppe and M. Marrocco, Phys. Rev. Lett. 76, 900, (1996).

[2] F. De Martini, O. Jedrkiewicz and P. Mataloni, Journal of Modern Optics, 44, 2053, (1997).

[3] F. De Martini and M. Giangrasso, Appl. Phys. B 60, S-49, (1995). F. De Martini and M. Giangrasso ”Microcavity Quantum Electrodynamics” in AmazingLight, ed. by R. Y. Chiao, (Springer, New York 1996).

[4] R.B. Dicke, Phys. Rev. 93, 99 (1954). L. Allen and J. Eberly, Optical Resonance and Two-Level Atoms (Wiley, New York, 1975). M Gross
and S. Haroche, Phys. Reports 93, 301 (1982). The condition of strong atom-field coupling is not met in the planar microcavity and this implies a small value of the product of the molecular decoherence time by the Rabi frequency, $T_2\Omega \ll 1$. As a consequence, the superradiant time phenomenology accounted for in the present work only relates to the behaviour of the diagonal elements of the density matrix of the active dipoles.

[5] F. De Martini, M. Marrocco and D. Murra, Phys. Rev. Lett. 65, 1853 (1991). A. Aiello, F. De Martini, M. Marrocco and P. Mataloni, Opt. Lett. 20, 1492 (1995). K. Ujihara, Jpn. J. Appl. Phys. 30, L901 (1991).

[6] P. Mataloni, O. Jedrkiewicz and F. De Martini, Phys. Lett. A 243, 270, (1998).

[7] F. De Martini, O. Jedrkiewicz and P. Mataloni, Journal of Nonlinear Optical Physics and Materials 7, 121 (1998).

[8] S. Ciancaleoni, P. Mataloni, O. Jedrkiewicz and F. De Martini, Journ. Opt. Soc. of America, B14, 1556 (1997).

[9] F. De Martini, M. Marrocco, P. Mataloni, L. Crescentini and R. Loudon, Phys. Rev. A, 43, 2480 (1991).

[10] R. Loudon, *The Quantum theory of Light* (Clarendon, Oxford, 1983), Chap. 5.

[11] F. De Martini and S. Di Fonzo, Europhys. Lett. 10, 123 (1989). The spatial effect we found may be interpreted as a kind of "Bose condensation".
FIGURE CAPTIONS

Fig. 1 - Optical configurations A) and B) of the Hanbury-Brown-Twiss interferometers. In both configurations the couples of equal single photon detectors are connected to a Multichannel Analyzer (MCA) through a Time-to-Amplitude Converter (TAC).

Fig. 2 - Two photon correlation function $F(\tau)$, in semilog scale, as function of the delay $\tau$ between the emitted photons for different relative values of the transverse inter-dipole distance: $R/\ell_c = 0.33 (\triangle)$, 2.9 (○), and 7.2 (*). The dashed straight lines represent the corresponding time decays evaluated by a quantum analysis. Inset: experimental normalized MCA distributions $F(\tau)$ for $R/\ell_c = 0.33$ (continuous line) and $R/\ell_c = 7.2$ (dotted line).

Fig. 3 - Two photon partition probabilities $P_{k,k'}(1,1)$ and $P_{k,k'}(2,0)$ over the two channels $k$, $k'$, detected at $\tau \approx 0$ as function of the relative transverse inter-dipole distance $R/\ell_c$. Note that the common quantum Bose - Einstein value of the probabilities and the two different classical Maxwell-Boltmann (M-B) values are reached correspondingly for $R/\ell_c \ll 1$ and $R/\ell_c \gg 1$. 