GENERATION OF ANTIBUNCHED LIGHT BY EXCITED MOLECULES IN A MICROCAVITY TRAP

F. De Martini, G. Di Giuseppe and M. Marrocco (*)
Dipartimento di Fisica and Istituto Nazionale di Fisica Nucleare
Universita' di Roma "La Sapienza", 00185 Italy

Abstract

The active microcavity is adopted as an efficient source of non-classical light. By this device, excited by 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. The process of adiabatic recycling within a multi-step Franck-Condon molecular optical-pumping mechanism, characterized in our case by a quantum efficiency very close to one, implies a pump self-regularization process leading to a striking n-squeezing effect. By a replication of the basic single-atom excitation process a beam of quantum photon $|n\rangle$-states (Fock states) can be created. The new process represents a significant advance in the modern fields of basic quantum-mechanical investigation, quantum communication and quantum cryptography

1 Introduction

The generation of non-classical 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, on a more technological perspective, the realization of a reliable source of this kind of radiation is today considered to be an essential feature of any realistic advanced program involving quantum cryptographic communication and, possibly in the future, quantum computation [1,2]. For this purpose the method of pump self-regularization has been adopted in the past within a few dynamical processes to provide the 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 [4] and the Rabi dynamics in resonant-fluorescence with excitation of single atoms in a beam, in a trap or in a solid host [5,6,7]. 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, of the hard problem of discriminating a very weak beam in the presence of a strong one at the *same* wavelength and, most important, of 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, efficient single-molecule

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 non-classical single-photon state that can be transformed into a quantum Fock |n>-state generator.

2 Experimental setup

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 single longitudinal-mode microcavity, with relevant dimension $d = m\lambda/2$, m=1, finesse f=1600, 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 be localized within a small volume $V = d \bullet s_p$ about equal to λ^3 at the intersection of the cavity active layer with the focal region of a 3 cm f-l lens collecting the excitation from a pulsed laser beam operating at λ_p. In the best configuration the device was excited by a collision-pulse-mode-locked (CPM) laser emitting at λ_p = 615 nm a sequence of equal pulses, referred to as "δt-pulses", with duration $\delta t = 0.1$ps, energy $\in$=0.12 nJ, rate $\nu = (1/\Delta t)$=100 Mhz. The experiment was also carried out, successfully but with far more critical requirements for the parameters δt, $\in$, at λ_p=532 nm, with a δt= 5 nsec, ν=20 Hz, pulsed beam SHG by a Nd-Yag Q-switched laser. The selected active system was a molecular solution in ethilene-glycol, a very viscous solvent at T=300°K , with concentration in the range $\rho = 10^{12} \div 10^{18}$ cm^{-3}, absorption cross-section $\sigma_p(\lambda_p) = 2.10^{16}$ cm^2 and free-space SpE time $(T_1)_0 \equiv 1/\Gamma_0 \approx 4$ nsec at the emission λ=702 nm at which the microcavity is tuned. Furthermore, very important,the selected molecule had a singlet four-level optical pumping quantum efficiency η very close to one [9]. With a calibrated ρ and 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 a 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 over two counter-propagating plane-wave modes with vectors **k** and **k'**= -**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* pump photon extracted from the laser beam, i.e., with poissonian statistics, at λ_p is re-emitted at a *different* λ over **k** or **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 4-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 $\xi_1 \approx \xi_2 \approx 0.12$, 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 this experiment, 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 **k, k'**, as shown by Fig. 1.

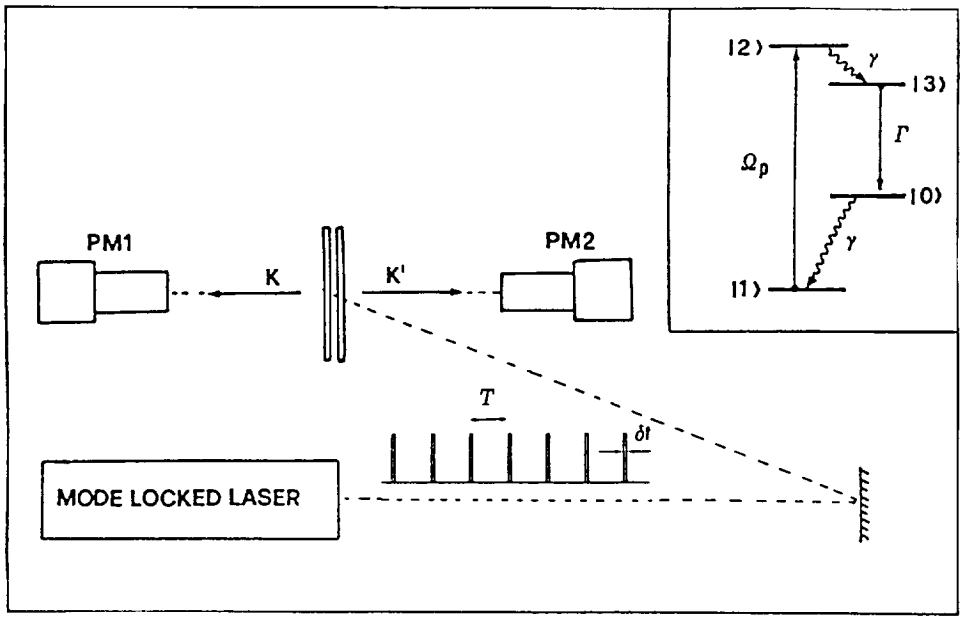

Fig. 1. Collision-Pulse-Mode-Locked laser-excited microcavity and Hanbury Brown-Twiss apparatus.

These ones 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 to interpret the microcavity as a new kind of *active beam-splitter.*

3 Quantum description of the antibunching process

Let's 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 **k**, **k'** with corresponding mutually commuting operators: $\hat{a} \equiv \hat{a}_{\mathbf{k}}$, $\hat{a}' \equiv \hat{a}_{\mathbf{k}'}$ [11].

The normal-ordered Hamiltonian of the system is:

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

being $\hat{o}$ the single-mode pump field operator, K_p , $K \propto \sqrt{\eta}$ appropriate coupling parameters proportional to corresponding Rabi frequencies Ω_p, Ω, $\eta \approx 3$ the microcavity field-enhancing factor, $\hat{\pi}_{ij} \equiv |i\rangle\langle j|$ (i,j = 0 to 3) the transition operators relative to the 4 -level system modelling the relevant features of the Franck-Condon dynamics of the single molecule. The system 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 only connected by a roto-vibrational fast relaxation process via a single coupling parameter: $\gamma \equiv 1/T_2 \approx 5.10^{12}$ sec^{-1} [12]. The transition operators are: $\hat{\pi}^- \equiv |0\rangle\langle 3|$, $(\hat{\pi}')^- \equiv |1\rangle\langle 2|$ 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, quasi-exponential 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: $\gamma >> \Gamma$. 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 [14]. For instance, the intensity $\langle : \hat{I}(t') : \rangle$ radiated after excitation by a sequence of equal δt-pulses, with $\delta t << \Gamma^{-1}$ and time interval $\Delta t \equiv \nu^{-1}$:

$$\langle \hat{E}^-(z, t')\, \hat{E}^+(z, t') \rangle = K \bullet \mathrm{rep}_T \langle \hat{\pi}^+(t)\hat{\pi}^-(t) \rangle, \quad \mathrm{rep}_T\, u(t) \equiv \sum_{-\infty}^{+\infty}{}_n\, u(t - n\Delta t) \quad [15]. \qquad (2)$$

For $t > \delta t$ is found:

$$\langle \hat{\pi}^+(t)\hat{\pi}^-(t) \rangle = A\{1 - \exp(-3\gamma\delta t/2)\cos(\lambda\,\delta t) + B\exp(-3\gamma\delta t/2)\sin(\lambda\,\delta t)\}\exp(-\Gamma t), \qquad (3)$$

with: $\lambda = [|\Omega_p|^2 - (\gamma/2)^2]^{1/2}$, $A \approx -(3/2)[\gamma|\Omega_p|/(|\Omega_p|^2 + 2\gamma^2)]^2$, $B \approx (|\Omega_p|^2 - 5\gamma^2/2)/(3\gamma\lambda)$ for $\Gamma << \gamma$. 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: $\Omega_p \bullet T_2 \approx [6\,\sigma_p \in /(\gamma\,\delta t\, s_p\, \hbar\omega_p)]^{1/2} > 1$, $\Omega_p \bullet \delta t \approx \pi$.

δt-pulse is a π-pulse for the overall dynamics, since: $\Omega_p \bullet T_2 \approx [6\,\sigma_p \in /(\gamma\,\delta t\, s_p\, \hbar\omega_p)]^{1/2} > 1$, $\Omega_p \bullet \delta t \approx \pi$. Then, if a single molecule interacts with that pulse, the excitation does not have time to cycle more than once within the 4-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 with a Poisson-type multiple emission [9]. The above analysis is completed by the evaluation of the degree of second-order coherence:

$$g^{(2)}(\tau) = <:\hat{I}(t')\hat{I}(t'+\tau):>/[<:\hat{I}(t'):>]^2. \tag{4}$$

This relevant quantity is evaluated, as usual, by first expressing the emission intensity average $<\hat{\pi}^+(t+\tau)\hat{\pi}^-(t+\tau)>$ 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 $|3>$, $|0>$, we may write the intensity average in the simple form:

$$<\hat{\pi}^+(t+\tau)\hat{\pi}^-(t+\tau)> = \beta_1(\tau) + \beta_2(\tau) \bullet <\hat{\pi}^+(t)\hat{\pi}^-(t)>, \tag{5}$$

where $\beta_1(\tau)$, $\beta_2(\tau)$ are evaluated by solving the master equation accounting for the emission process. This leads to a straightforward evaluation of $g^{(2)}(\tau)$. This quantity is given here for a 4-level molecule, for $\tau < \Delta t$ and for two extreme δt-pulse excitation conditions, $\delta t << 1/\Gamma$:

(a) excitation by a single δt-pulse: : $g^{(2)}(\tau) = [\beta_1(\tau)/\beta_1(\infty)] = [1 - \exp(-\Gamma\tau)]$

(b) excitation by a sequence of δt-pulses, rate Δt^{-1} : $g^{(2)}(\tau) = \Gamma\Delta t \cdot [1 - \exp(-\Gamma\tau)]/[1 - \exp(-\Gamma\Delta t)]$;

$\Gamma\Delta t > 1$. We see that in both cases is $g^{(2)}(0) = 0$, 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:

$$\alpha(t, \Delta t_g) = \frac{\langle p_c(t, \Delta t_g)\rangle}{\langle p_1(t, \Delta t_g)\rangle \cdot \langle p_2(t, \Delta t_g)\rangle} \tag{6}$$

For single mode excitation of the beam splitter, $\Delta t_g << T_1$, we obtain by quantum theory [3]:

$$\alpha(\bar{n}) \equiv \alpha\left(0, \Delta t_g\right) = \frac{\mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_1 \hat{a}_1^{\dagger}\hat{a}_1\right)\right]\left[1-\exp\left(-\xi_2 \hat{a}_2^{\dagger}\hat{a}_2\right)\right]\right\}}{\mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_1 \hat{a}_1^{\dagger}\hat{a}_1\right)\right]\right\} \bullet \mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_2 \hat{a}_2^{\dagger}\hat{a}_2\right)\right]\right\}} \tag{7}$$

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*: $P_n = \bar{n}^n / (1+\bar{n})^{1+n}$ $\alpha = [2+\xi_1 T' \bar{n}+\xi_2 R' \bar{n}]/[1+\xi_1 T' \bar{n}+\xi_2 R' \bar{n}]$

2) *Coherent*: $P_n = \frac{\bar{n}^n}{n!}\exp(-\bar{n})$ $\alpha = 1$

3) *Antibunched*: $P_n = \delta_{n,\bar{n}}$ $\alpha = [1+(1-\xi_1 T'-\xi_2 R')^{\bar{n}} -(1-\xi_1 T')^{\bar{n}}-(1-\xi_2 R')^{\bar{n}}] \bullet$
$\bullet\{[1-(1-\xi_1 T')^{\bar{n}}][1-(1-\xi_2 R')^{\bar{n}}\}^{-1}$

being $R'=|r|^2$, $T'=|t|^2$ the optical parameters of the (loss-less) beam splitter and $\bar{n}$ 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: $g^{(2)}(0) = [\alpha - B(\bar{n})]\bullet[A(\bar{n})]^{-1}$, where:

$$A(\bar{n}) = \frac{\mathrm{Tr}\left[\hat{\rho}\hat{a}_1^{\dagger}\hat{a}_1\right] \bullet \mathrm{Tr}\left[\hat{\rho}\hat{a}_2^{\dagger}\hat{a}_2\right]}{\mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_1 \hat{a}_1^{\dagger}\hat{a}_1\right)\right]\right\} \bullet \mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_2 \hat{a}_2^{\dagger}\hat{a}_2\right)\right]\right\}} \tag{8}$$

and,

$$B(\bar{n}) = \frac{\mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_1 \hat{a}_1^{\dagger}\hat{a}_1\right)\right]\left[1-\exp\left(-\xi_2 \hat{a}_2^{\dagger}\hat{a}_2\right)\right] - \hat{a}_1^{\dagger}\hat{a}_1\hat{a}_2^{\dagger}\hat{a}_2\right\}}{\mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_1 \hat{a}_1^{\dagger}\hat{a}_1\right)\right]\right\} \bullet \mathrm{Tr}\left\{\hat{\rho}\hat{N}\left[1-\exp\left(-\xi_2 \hat{a}_2^{\dagger}\hat{a}_2\right)\right]\right\}} \tag{9}$$

According to the theory, for n=1 is: $g^{(2)}(0) = \alpha = 0$.

4 Experimental result

The parameter α is plotted in Fig. 2 for the three cases Vs. $\bar{n}$ and the molecular $\rho \propto \bar{n}$, for our experimental conditions.

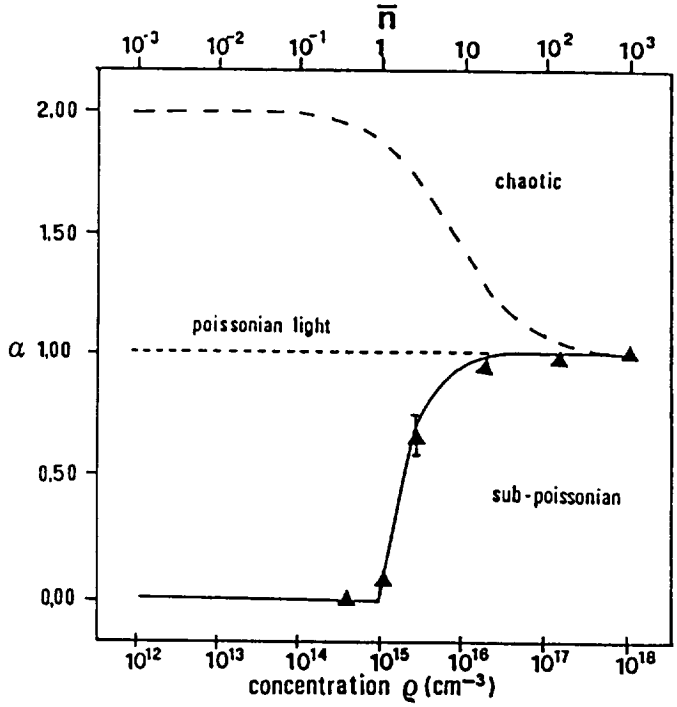

Fig. 2. Coincidence parameter α ($\bar{n}$) as function of the number of photons emitted after each exitation pulse and of the molecular concentration. The time-gate of the HBT apparatus was: Δt_g = 1 nsec.

Note in Fig. 2 the good experimental verification for $\bar{n}>1$ of the theoretical curve expressing α ($\bar{n}$) in the sub-poissonian condition, viz., implying the pure n-state distribution: $P_n = \delta_{n,\bar{n}}$. These results of the HBT experiment show that an increasing sub-Poisson character of the output radiation is gradually established for ρ varying over two-order of magnitude, leading for $\rho \approx 7 \bullet 10^{14}$ cm^{-3} to the striking figure $\alpha = g^{(2)}(0) = 0$ for $n=\bar{n}=1$. This last result has been obtained at T=300°K with a 50%-50% beam-splitter within a run involving a number of counts equal to $1.5 \bullet 10^4$ 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.

5 Conclusion

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 a single-photon per pulse, with a quasi *deterministic* generation of a quantum radiation state, at repetition rates as high as 100 Mhz and with a quantum efficiency close to one. This result leads to a still more important consequence. The single-molecule excitation process could be straigthforwardly

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>>1): $\tau_t=l_t/c\approx 2(\lambda/c)\sqrt{f}<<\tau_c\approx(\lambda/c)f$, then the indistinguishable emitted n single-photons do belong to the *same* space-time extension of the output field-mode, i.e., they form a quantum $|n\rangle$-state [20]. The experimental realization of these conditions would certainly determine a new exciting endeavour within the quantum optics community. The preliminary results of our investigation in this direction are quite encouraging. We acknowledge useful discussions with P. Milonni, J. Franson and Y. Shih ■

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(*) Present address: Max-Planck Institut fur Quantenoptik, Garching D-85748, Germany.

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generated at that active center is directed, with estimated *efficiency* ϕ >96% , over either one of the two output modes **k, k'** in spite of the limited optical confinement provided by the Bragg reflectors at large emission angles respect to **z**, viz. to the direction of **k** (Fig.1). When needed, the use of one totally reflecting microcavity mirror, R=100% allowed the excitation of *only one* external mode, **k**. Under the given experimental conditions justifying, for $\Omega_p \bullet T_2 > 1$, the adoption of a quantum Rabi dynamics, and the efficiencies $\eta \approx 1$, $\phi \approx 1$, it is assumed that the result $\alpha = g^{(2)}(0) = 0$ implies a single-molecule excitation in the active region. For a very preliminary account of the present work, involving long excitation pulses, $\delta t \approx 5 \bullet 10^{-9}$ s. cfr: M. Marrocco and F. De Martini, in: *Quantum Interferometry,* ed. by F. De Martini, G. Denardo and A. Zeilinger (World Scientific, London, 1994).

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