Transverse Quantum Correlations in the Active Microscopic Cavity

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A microcavity technique is adopted to investigate, we believe for the first time, the process of *transverse* interatom quantum correlations taking place after photon emission, according to the plane-wave QED photon-delocalization model. We find that the QED model is indeed verified but that it is generally limited by the nonideal cavity properties. The concept introduced here of "transverse quantum-correlation length" is of fundamental relevance to all kinds of Casimir-type vacuum-confinement processes, including "anomalous" QED effects in spontaneous emission. It also sets a limitation on the performance of the "thresholdless laser," a device of actual scientific and technological interest.

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Anomalous vacuum-confinement dynamical effects in the context of optical spontaneous emission (SpE) and stimulated emission (StE) by an active microscopic cavity (microcavity) have been reported recently.[1,2] It was shown that the low-dimensional modal structure of the field in the microcavity determines the striking effects on the atomic SpE lifetime and a peculiar "thresholdless-laser" effect.[2] This last effect, due to the virtual elimination of the predominant source of cavity damping related to mode competition, realizes for the first time in an optical system the deterministic one-degree-of-freedom limit of the statistical "mode reservoir" in StE field-atom interactions.[2,3] The unusual properties of the zero-threshold microlaser have recently attracted a great deal of interest due to its technological importance in the field of integrated active semiconductor devices.[4]

While reporting in Ref. 2 the first realization of this device, we emphasized there the gain properties of the system in which the excited atoms, being actively coupled to the same cavity mode, cooperate in StE due to the QED photon-delocalization model. According to this model, first introduced by Einstein, after atomic deexcitation the photon field undergoes a plane-wave delocalization with definite momentum $\mathbf{p}=\hbar\mathbf{k}$.[2] While this is certainly true in idealized conditions, as we shall see, in the present work we demonstrate that the cooperation effect is actually limited by the damping properties of the Fabry-Pérot (FP) microcavity, i.e., by its limited finesse f.[5] Furthermore, we find that this inhibition effect is increasingly larger for increasing quantum confinement, reaching its maximum at the minimum "SpE-enhancement" cavity spacing, $d=N\bar{d}$, $\bar{d}\equiv\lambda/2$, i.e., for $N=1$.[2] In our experiment the extent of the "transverse" (i.e., acting along a direction orthogonal to the propagation $\mathbf{k}$ vector) interatom quantum correlations is determined by investigating the StE coupling established between two equal localized sets of excited molecules placed in two spots, 1 and 2, in the microcavity and coupled to the same cavity-allowed "plane-wave" mode with $\mathbf{k}$ vector orthogonal to the cavity plane mirrors (A,B,

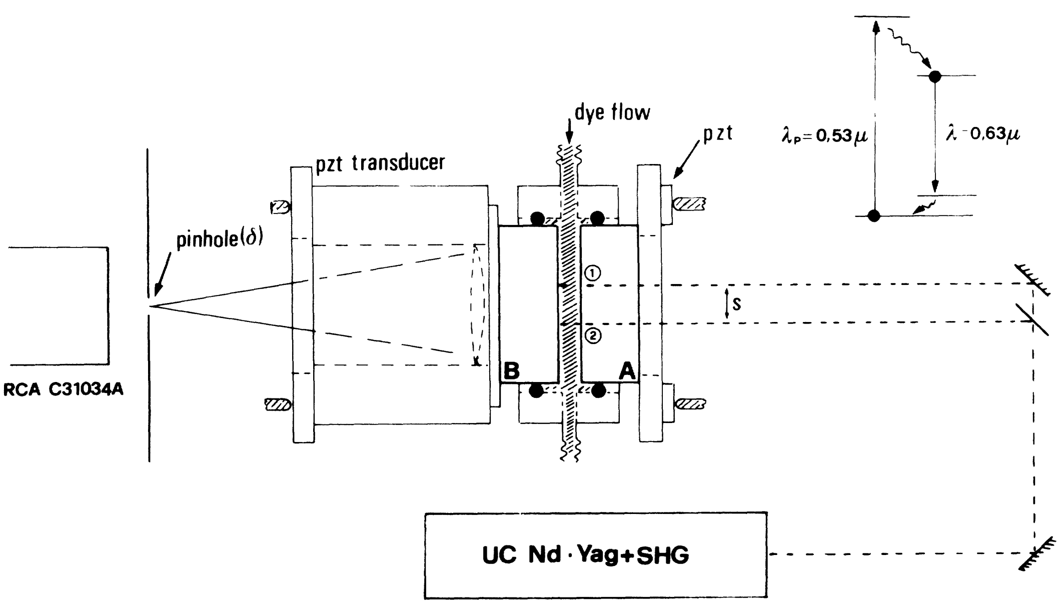

FIG. 1. Schematic diagram of the experimental apparatus.

Fig. 1). This problem, which generally involves a peculiar transverse relativistic-retardation process, should be taken as a basic one in any laser StE calculation and, to our knowledge, has not been investigated before.[3,6] Take, as said, two equal and dynamically equivalent cylindrical "microlasers" 1 and 2, with diameters δ and placed at an adjustable mutual distance s in the microcavity active plane. In our experiment this was obtained by focusing on two $\delta \approx 30$ μm spots on the active plane, by use of a common lens (focal length, 25 cm), two TEM_{00} single-mode pump beams ($\lambda_p = 0.53$ μm) whose noncollinearity before focusing was micrometrically controlled. The problem tackled in this work may be reformulated by the following question: "To what extent does a StE photon emitted over the common **k** mode by one microlaser determine the gain of the other microlaser, in spite of a macroscopic distance s, taken orthogonally to **k** and externally imposed on the two lasers?" According to QED, since photon delocalization is expected after emission over the transverse extension of the mode, full interlaser correlation is also expected in spite of the peculiar topological configuration of the experiment.[7]

Let us analyze the problem by considering the single-mode case $d = \bar{d}$, for simplicity. Let m_1, m_2 be the numbers of photons emitted by microlasers 1 and 2, respectively, within the coherence time $t_c = \lambda/(v\Delta\lambda)$ over the common **k** mode ($\Delta\lambda \equiv$ bandwidth of the detected radiation). The time-evolution equation for m_1 is given in the form $dm_1/dt = G(1 + m_1 + \alpha m_2)$, where the "degree of correlation," $\alpha = \alpha(s,d)$, $0 \le \alpha \le 1$, represents interlaser coupling. The extreme values taken by α in its existence range correspond to full laser independence and to full correlation, respectively. An identical equation holds for m_2 by interchanging indices. At last, the overall emitted photon number $m \equiv m_1 + m_2$, relative to the condition $\alpha = 1$, is related to $\alpha(s)$ through

$$f(s,d) \equiv \frac{m(s,d)}{m(0,d)} = \frac{2\sinh[G(\alpha+1)]\exp[G(\alpha-1)/2]}{(\alpha+1)\sinh(2G)}, \qquad (1)$$

where $G \equiv g d I_p$ is the low-signal gain proportional to the microlaser pump intensity I_p and to d, in first approximation.[6] Note that the overall output gain is strongly dependent on α, as it almost *doubles* in the case of $\alpha = 1$. The measure of gain as a function of s and d is precisely the method we adopt to investigate the quantum-correlation process.

Before reporting the experimental results, let us give some details on the apparatus. A negative-branch, unstable-cavity, Q-switched neodymium-doped yttrium-aluminum-garnet laser equipped with a second-harmonic generator provided TEM_{00} single-mode pulses at λ_p with $\tau = 10$ nsec duration to pump the two microlasers upon focusing in the microcavity active plane. The mutual polarizations of the pump beams were taken at 90° in order to avoid field-interference effects within the pumping process. The pump intensity was kept at a level such that no laser saturation was detected, as shown by the slope of the gain curves in Fig. 2.[8] The cavity was similar to the one reported in Ref. 2. Mirror A was transparent to λ_p and reflected $R_1 = 99.9\%$ of the microlaser light at $\lambda = 6328$ Å, the detected wavelength. Mirror B was $R_2 \approx 98\%$ reflecting for λ and λ_p. A flow of a 10^{-2} ethanol solution of DCM dye was maintained between the mirrors. The cavity zeroth-order **k** mode was focused by a 30-cm focal-length lens, with lens aperture equal to 10 mm, onto a pinhole of diameter $\delta' = 10$ μm. The StE light reaching the phototube (RCA C31034A-02) was filtered by a $\Delta\lambda = 8$ Å interference filter centered at λ. The signals were processed by a computer-interfaced LeCroy 9400 digital oscilloscope.

The plots of the output-radiation intensity I emitted from the microcavity versus the pump intensity for $d = \bar{d}$, $5\bar{d}$, and $10\bar{d}$ are reported in Fig. 2, for various s values. The I_p scales for the three cases are renormalized to compensate for the effect of different d in determining $G(dI_p)$. A progressive loss of intermicrolaser correlation for increasing distance s is shown in Fig. 2 by the progressive departure of the gain curves from the full-coupling curve (obtained with $s = 0$) toward the curve of complete decoupling, the dashed one in Fig. 2. This

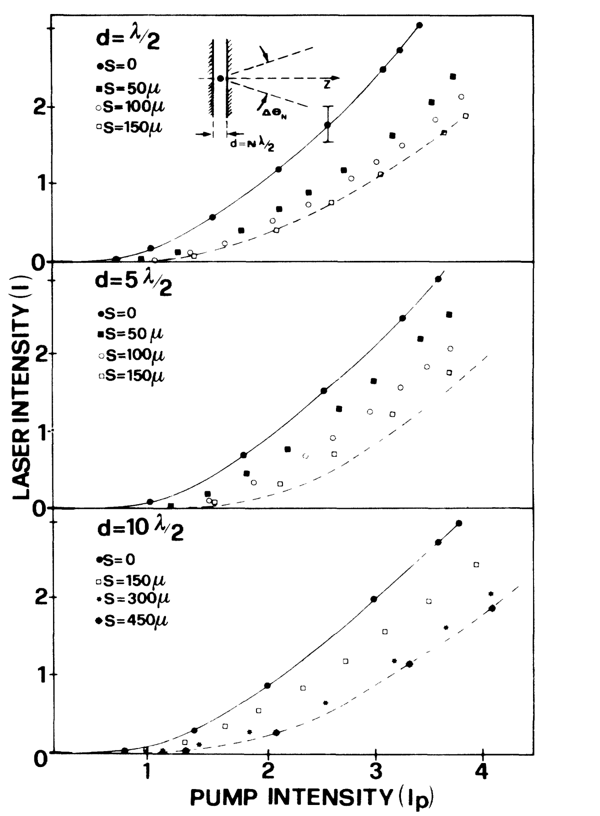

FIG. 2. Gain curves showing the loss of quantum correlation with mutual distance between two identical microlasers excited in a microscopic cavity (arbitrary and normalized scales for I and I_p).

curve is obtained by doubling the values of I and I_p related by the full-coupling curve. Figure 2 also shows that the effect of correlation loss is increasingly less pronounced for increasing $d > \bar{d}$, asymptotically approaching (i.e., for a macroscopic cavity, $d \gg \bar{d}$) the general behavior $\alpha(s) \approx 1$ expected according to standard theory. The measurement of $I(I_p,s)$ leads to the determination of $\alpha(s)$, through Eq. (1). The α curves for $d = \bar{d}$, $5\bar{d}$, and $10\bar{d}$, shown in Fig. 3 together with related best-fit Gaussian plots, reproduce the relevant correlation-increasing behavior for increasing $d = N\bar{d}$.

An approximate, simplified explanation of this behavior may be given as follows.[9,10] Consider the microcavity forward mode, i.e., with average **k** vector orthogonal to the mirrors, and corresponding to $N \geq 1$. For an active FP cavity with finesse $f = \pi R^{1/2}/(1-R)$, $R^2 \equiv R_1 R_2$, this mode may be considered as a superposition of plane waves with a **k**-space distribution assigned by the FP "transfer function," i.e., by the Airy function $Y \propto [1 + (2f/\pi)^2 \sin^2\psi]^{-1}$ and by the related expression of the FP interference phase $\psi = \pi N \cos\Theta$, as a function of the emission angle Θ.[5] The full width at half maximum (FWHM) of the **k**-vector distribution is found: $\Delta\Theta_N = 2(fN)^{-1/2}$ (Fig. 2, inset). The superposition of plane waves, over each of which the forward-emitted photons are delocalized, determines, according to field-interference considerations, a limitation of the transverse coherence length over which StE correlations can effectively take place.[10] This process may be accounted for in simple terms by the following argument. The extent of the photon-gas Gibbs phase space corresponding to the cavity forward mode is expressed, for a rectangular spatial cross section $\Delta x \Delta y$, by $\Delta x \Delta y \Delta z \Delta p_x \Delta p_y \times \Delta p_z = h^3$, where $\Delta z, \Delta p_z$ refer to the time coordinate. This of course may be interpreted as expressing the minimum-uncertainty application of Heisenberg's principle to the tridimensional dynamics of the photon particles localized within the coherence extent of the mode. By writing $\Delta p_x \Delta p_y = (\hbar k \Delta\Theta_N/2)^2$, we finally obtain, for cylindrical symmetry, the expression for the *transverse quantum-correlation length*, $l_N = 2\lambda(fN)^{1/2}$. Then, two cylindrical microlasers with diameter δ, sharing a common FP cavity mode, can be coupled by StE if $l_N > \delta$. This relation leads to further interesting insight into the dynamics of the process. In fact, if $\delta < l_N$ the microcavity-allowed **k**-space distribution is *sharper* than the one required in the nonconfinement condition by Fresnel diffraction from the circular ends of each active microlaser, or $\Delta\Theta_N < \Delta\Theta_D \approx \lambda/\delta$. Since in this case the diffraction process is made ineffective in determining the output **k** distribution, we may say that there the microcavity *inhibits diffraction*. The backreaction of the field to this anomalous condition consists of a kind of a "coupling halo," a cylindrical region of thickness $\frac{1}{2}(l_N - \delta)$ surrounding the microlasers in which quantum correlations can take place. We refer to this condition as the *cavity regime*. On the other hand, if $l_N < \delta$ no StE correlation in the transverse direction is possible outside the active regions: Here, diffraction inhibits external correlations. This identifies the *diffraction regime*. Since in this regime no transverse correlation takes place over radial distances $l > l_N$, the maximum number of StE-interacting atoms is $\bar{n} = \frac{1}{4}\pi\eta_N d l_N^2 = \frac{1}{2}\pi\eta_N f N^2 \lambda^3$, η_N being the fraction of the volume density of excited atoms, η, corresponding to emission over the forward mode (in microcavities, $\eta_N < \eta/N$).

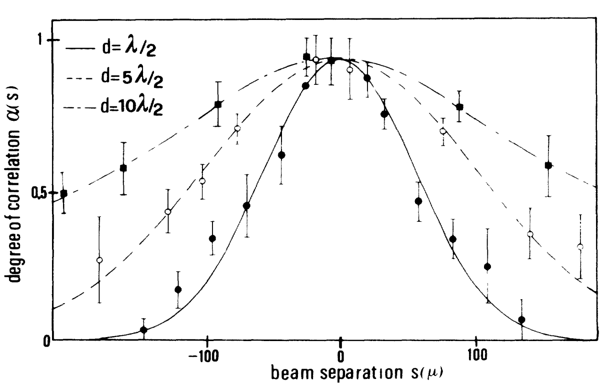

FIG. 3. Degree of Bose-Einstein correlation $\alpha(s)$ as a function of the intermicrolaser distance s for microcavity spacings $d = \bar{d} \equiv \lambda/2$, $5\bar{d}$, and $10\bar{d}$.

The above considerations lead us to conclude that in open space, viz., outside a *mode-selective* cavity, it is impossible to detect transverse field-delocalization effects using our StE technique involving active spots separated by $l > \lambda$: There, in fact, the diffraction regime is always realized. In fact, by our arguments we find $l_N \approx \lambda$ for two isolated atoms cooperating in open space, in the absence of external fields, in transverse photon emission, viz., emitting along a direction orthogonal to any plane to which the two atoms belong.[10] On the other hand, the use of an ideal cavity with infinite finesse leads to a single-plane-wave structure for the forward mode, $\Delta\Theta_N = 0$, and to quantum correlations extending over the full transverse extent of that mode: $l_N = \infty$.[10] In addition to that, and very important, since the transverse quantum-correlation length identifies a region of spatial coherence for the quantum field, including the *vacuum field*, l_N identifies the transverse extent of the modes (and of the cavity mirrors) that are effectively involved in the dynamics of spontaneous emission from one excited atom.[1] The microcavity regime is demonstrated experimentally in Fig. 3 for increasing $N = 2d/\lambda$. The value of f ($f \approx 170$) has been determined by direct measurement of the angular distribution of the output intensity, $\Delta\Theta_N$, according to our theory.[11] This one has been further substantiated by the results of a similar experiment involving a relatively long microcavity, $N = 10^3$, terminated by simple glass windows, $f \approx 0.3$.

In summary, we have investigated by a new technique the rather unexplored condition of transverse nonlocality of the electromagnetic field. Consequently, our work may stimulate future endeavors of fundamental scientific relevance. For instance, since nonlocality is closely related to quantum nonseparability, it may suggest the realization of a new kind of Einstein-Podolsky-Rosen-type experiment, opening then a new field of fruitful quantum-mechanical speculation.[12]

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[1]F. De Martini and G. Innocenti, in *Quantum Optics IV*, edited by J. Harvey and F. Walls (Springer-Verlag, Berlin, 1986); F. De Martini, G. Innocenti, G. Jacobovitz, and P. Mataloni, Phys. Rev. Lett. **59**, 2955 (1987); W. Jhe, A. Anderson, E. Hinds, D. Meschede, L. Moi, and S. Haroche, Phys. Rev. Lett. **58**, 666 (1987).

[2]F. De Martini and J. R. Jacobovitz, Phys. Rev. Lett. **60**, 1711 (1988); F. De Martini, Phys. Scr. **21**, 58 (1988). The microcavity single-mode concept is an approximate one. In fact, in our cavity, weakly excited, nonresonant radial modes propagating in directions parallel to the mirrors are also allowed. Casimir-type extreme confinement is realized for field confinement over a space-time distance of the order of the field's de Broglie wavelength $\lambda_D = \lambda$. The photon-delocalization model is at the basis of the modern quantum theory of the Doppler effect in atomic spectroscopy, cf. R. Loudon, *The Quantum Theory of Light* (Clarendon, Oxford, 1983), Chap. 2. For the old model, cf. A. Einstein Phys. Z. **18**, 121 (1917).

[3]M. Sargent, M. O. Scully, and W. E. Lamb, *Laser Physics* (Addison Wesley, New York, 1974), Chap. 14.

[4]Y. Yamamoto, in Proceedings of the International Quantum-Electronics Conference, Anaheim, California, May 1990 (to be published).

[5]M. Born and E. Wolf, *Principles of Optics* (Macmillan, New York, 1964), Chap. 8.

[6]A. Yariv, *Quantum Electronics* (Wiley, New York, 1967), Chap. 15; F. Shafer and W. Schmidt, Phys. Lett. **9**, 306 (1966). In our experiment, $t_c \approx 1.7 \times 10^{-12}$ sec. The retardation time between microlasers 100 μm apart is $t_r \approx 5 \times 10^{-13}$ sec. Then, $t_r \ll \tau$, the pump-pulse time duration. This implies that the reduction of StE correlation for $s > 0$ cannot be attributed to trivial retardation in the radial direction. According to the QED photon-delocalization model, the laser theories developed so far appear to imply the tacit assumption that all excited atoms belonging to the transverse section of the active medium always cooperate in StE. The present work shows that, while this may be true for long-cavity, high-Q lasers, it *is not* true in general, in particular for active microcavities.

[7]Note the peculiar field-retardation process taking place in our experiment. In an idealized situation, a photon emitted by microlaser 1 cannot interact with 2 in a time shorter than t_r as a StE correlation corresponds to an "information exchange" between lasers. Information transfer is certainly provided by photons belonging to the microcavity "radial" modes. However, StE effects taking place on radial modes cannot be detected by our lens-pinhole mode-selecting detector which is only sensitive to StE on the forward mode. For retardation in atom-radiation processes, cf. E. Fermi, Rev. Mod. Phys. **4**, 7 (1932); S. Kikuchi, Z. Phys. **66**, 8 (1930); M. Fierz, Helv. Phys. Acta **23**, 731 (1950); P. Milonni and P. Knight, Phys. Rev. A **10**, 4 (1974); V. P. Bykov, Usp. Fiz. Nauk **143**, 657 (1984) [Sov. Phys. Usp. **27**, 631 (1984)].

[8]Any StE-saturation process would tend to cancel the evidence of StE correlation by flattening down the $\alpha(s)$ curves of Fig. 3.

[9]A detailed mean-field theory based on the solution of collective Heisenberg equations for a superradiant medium will be reported elsewhere. N. Rehler and J. H. Eberly, Phys. Rev. A **3**, 1735 (1971); J. A. Stratton, *Electromagnetic Theory* (McGraw-Hill, New York, 1942), p. 256.

[10]An equivalent, rigorous approach for the determination of l_N is provided by Feynman-path interference considerations related to the coupling problem. There *each* path corresponds to a single deexcitation channel, viz., to a single **k** vector available for atom deexcitation. Exactly for this reason the "photon-localization" effect (or, $l_N < \infty$) found in the present work depends on the spatial-mode structure of the field coupled to the excited atom. This structure is determined by the external boundary conditions, e.g., by the presence or absence of a cavity. This approach is closely related to the one based on "diffraction," which is a typical field-interference process. R. P. Feynman and A. Hibbs, *Quantum Mechanics and Path Integrals* (McGraw-Hill, New York, 1965), Chap. 1; K. Huang, *Statistical Mechanics* (Wiley, New York, 1963), Chap. 9. With usual macroscopic cavities, $N \gg 1$ and $l_N \gg \lambda$: e.g., with $f = 100$, $d = 10$ cm, and $\lambda = 0.7$ μm we have $l_N \approx 7.5$ mm.

[11]Over short radial distances $\approx l_N$, the f values measured by the emitted-intensity angular distribution can be very high, as determined mostly by R. The small f values reported in works Refs. 1 and 2 were obtained by measuring the d dependence of the microcavity transmission using a large-diameter beam (>10 mm). There, large f-limiting effects due to imperfect mirror planarity were overwhelming. The results of the present work are consistent with our previous microcavity results. Note, for instance, that the data reported in Fig. 1 of Ref. 2 (De Martini and Jacobovitz) show that, for constant $I_p \propto$ (number of excited atoms) and for increasing $d = N\bar{d}$, the negative effect on the gain due to mode competition over $N' > N$ transverse and longitudinal cavity modes prevails over the positive effect due to the increase of $l_N \propto \sqrt{N}$. The present work shows that the optical cavity does not merely provide a generic "enhancement" of the atom-field interactions, as is generally believed. In spite of its simplicity, it is in fact a beautiful device of fundamental physical relevance as it appears to provide the only solution for extending the range of the interatom transverse quantum correlations from the microscopic to the *macroscopic* domain. The effect of f dependence of the degree of StE quantum cooperation should provide a hint for the conception of a new kind of nonlinear optical multistable device and of a new mode-locking scheme.

[12]M. Jammer, *The Philosophy of Quantum Mechanics* (Wiley, New York, 1974), Chap. 6; A. Einstein, B. Podolsky, and N. Rosen, Phys. Rev. **47**, 777 (1935). The problem investigated in the present paper is reminiscent of (and it is somewhat reverse to) the space-time transverse-localization argument expressed by Einstein at the 1927 Solvay Conference and dealing with relativistic causality in particle detection over a diffracted wave front: cf. N. Bohr, in *Albert Einstein Philosopher-Scientist*, edited by P. A. Schlipp (Cambridge Univ. Press, London, 1982); F. De Martini, in *Proceedings of the Niels Bohr Symposium, Rome, 1985* [Riv. Storia Sci. **2**, 557 (1985)].