Anomalous Spontaneous Emission Time in a Microscopic Optical Cavity

F. De Martini, G. Innocenti, G. R. Jacobovitz,[a] and P. Mataloni
Dipartimento di Fisica Università di Roma I, 00185 Roma, Italy
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We have realized total electromagnetic mode confinement in a microscopic optical, Casimir-type, cavity and detected the resonant change of the molecular fluorescence time under short-pulse excitation due to a spontaneous-atomic-decay enhancement-inhibition process (Purcell effect). This corresponds to the first realization in optics of the resonant coupling of atoms with a single mode of the radiation field.

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The problem of the interaction of atoms or molecules with the radiation field in its quantum ground state and in the presence of electromagnetic boundaries has attracted in the past a great deal of attention, both on the theoretical[1,2] and the experimental sides.[3-5] In recent times, spontaneous-emission (SE) enhancement-inhibition processes in macroscopic cavity structures have been investigated mostly in the microwave and infrared zones of the spectrum.[3] A very recent paper by Heinzen *et al.* deals with such an effect in the optical range, reporting evidence of the resonant behavior of the light intensity emitted by atoms placed in a large-size confocal resonator and detected in a "continuous-wave" (CW) condition.[4]

The present experiment is substantially different from that work because a "microscopic" optical cavity (or "microcavity," i.e., with size $d \leq \lambda$, the SE wavelength) is involved, and a very direct measurement of the atomic relaxation time T is performed by the detection of, for the first time in this kind of experiment, the spontaneously emitted radiation in the time domain.[5] The significance of these features may be appreciated if one considers that, in general, two entirely different resonant processes affect any SE light-intensity measurement in the presence of a cavity. First, the presence of a resonant structure necessarily affects the emission probability $W_\sigma = \sum_k^\sigma W_k$ of the set σ of the modes that are confined by the cavity itself. As a consequence, if one of these modes is probed out of the cavity, a resonant change of SE light intensity must be detected. This effect, corresponding to a light-intensity spatial redistribution, is observed even in the absence of any sizable resonant change of the average atomic spontaneous decay rate Γ.[4,6] A true resonant inhibition-enhancement effect on Γ [and then on the average SE rate $(1/T)$] may be realized experimentally if all (or at least a very large portion of the) radiation modes interacting with the atoms are confined within the cavity boundaries and are then affected by the resonance. For field confinement by plane boundaries, this condition can be realized only if the relevant linear dimension d of the resonator is of the order of the SE wavelength λ, a condition not easily realizable in the optical range.[3] Second, as shown by Heinzen *et al.*, an approximate alternative to this condition may be represented by the use of a macroscopic degenerate confocal cavity. However, in this case, where the resonant behavior of the SE light intensity is investigated in a CW condition and a full mode confinement is not achieved, the two competing effects overlap and may not be easily separated.[4] On the basis of these considerations and of the theoretical results given below, it appears that, in general, a direct nonambiguous test for the verification of the "real" effect, the Purcell effect,[1] is the detection of the resonant change of the SE lifetime T in the time domain and in a total-mode-confinement condition.[7]

Fulfilling this program, we have obtained the first realization of total mode confinement in a microcavity and the experimental evidence of the resonant behavior of the atomic spontaneous-emission lifetime in the time domain, under pulsed excitation.[5] A microscopic piezoelectrically tuned Fabry-Perot cavity (Fig. 1) is composed of two plane dielectric-coated mirrors (resonator finesse = 15). A steady flow of a 10^{-4} ethanol solution of tetraphenylnaphthacene dye (free space $T_0 = 1/A = 13$ nsec) was maintained between the mirrors. The optical pumping of the active medium was provided by second-harmonic generation at $\lambda_p = 0.53$ μm by an ultrastable self-injected neodymium-doped yttrium aluminum garnet laser with pulses of 2-nsec duration.[8] During the experiment, the intensity of the pumping beam was taken below the simulated emission threshold for $d < \lambda/2$. The pump beam was also well collimated and was injected into the cavity through one mirror (A) at a selectable angle ϑ_p, taken with respect to the cavity axis, in order to take advantage of the effect of "periodic optical pumping," a technique that we recently introduced and which is based on the self-interference of the pump beam on a reflecting mirror (B).[5,9] This technique allows one, by changes of ϑ_p, to position accurately the zones of pumping and then to control the location of the excited molecules in the intracavity space. By SE intensity measurements we found that $\vartheta_p = 48°$ was the angle corresponding to a selective molecular excitation located at $z = \lambda/4$

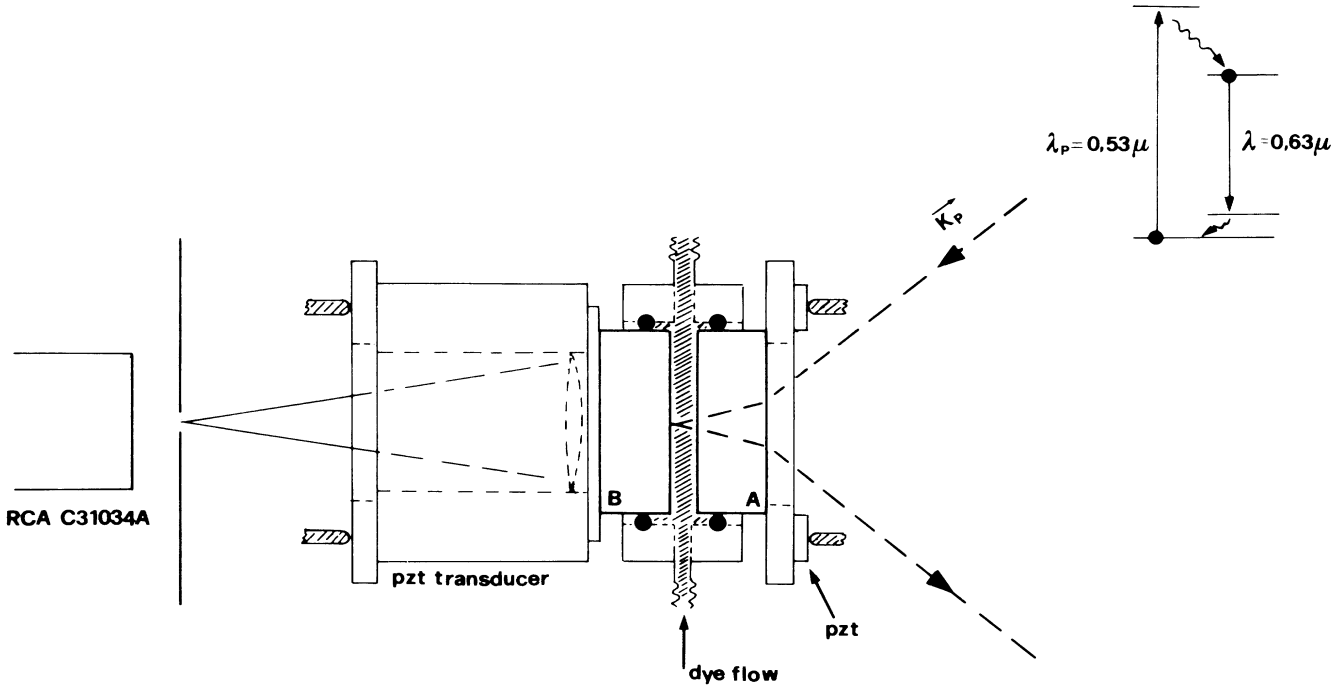

FIG. 1. Microscopic Fabry-Perot cavity (section). The directions of the incoming and reflected beams with the excitation wavelength $\lambda_p = 0.53$ μm are shown at the right-hand side.

from mirror B. This last mirror was 96% reflecting for the SE and the pump wavelengths (λ,λ_p), while mirror A was 98%-reflection coated at λ and antireflection coated at λ_p. The SE light emitted from the cavity was filtered by an interference filter centered at $\lambda = 6328$ Å (10-Å pass band), focused on the end of an optical fiber, and then recorded by a large-quantum-efficiency RCA C31034A photomultiplier (2-nsec rise time). The SE waveform data were computer processed by interfacing with a Le-Croy 8013A fast waveform digitizer (1.3-GHz bandwidth). The cavity alignment was maintained by means of two small disk-shaped piezoelectric transducers (PZR) while the cavity spacing was controlled by a Micro-Controle positioner (by 1000-Å steps) and, on a finer scale, by a large cylindrical PZT. We found that, fortunately, the cavity alignment for $d < \lambda$ was kept particularly stable by the surface tension of the dye solution.

The mirror spacing d could be varied over a distance ranging from about 1 mm down to a very small fraction of λ, thus realizing, for $d < \lambda/2$, the remarkable quantum-mechanical effects related to the field-mode elimination and the topological configuration of the Casimir effect.[10] Note that, for $d \lesssim \lambda$, the atoms in our cavity were forced to interact resonantly with a single radiation mode, thus realizing, for the first time at optical frequencies, a novel and quite interesting quantum-statistical situation.[11]

In Heisenberg's representation, the dynamics of the single-atom–field coupling is expressed, as usual by a Dicke Hamiltonian written in terms of the time-dependent atomic displacement operators $(\Pi^\dagger,\Pi)$ and field operators $(a_k^\dagger,a_k)$ belonging to the modes k.[12] By a Markov approximation, we obtain the evolution equations for the atom and the field belonging to the (single) mode h which is probed out of the cavity, for instance, along the cavity axis:

$$\frac{d}{dt}(\Pi^\dagger\Pi) = -\frac{d}{dt}\sum_k (a_k^\dagger a_k) \quad (\omega \cong \omega_k), \tag{1a}$$

$$\frac{d}{dt}(a_h^\dagger a_h) = -(g_h f_h)(\Pi^\dagger E_h^0 + \text{H.c.}) + \gamma_h(\Pi^\dagger\Pi). \tag{1b}$$

Equations similar to (1b) may be written for any deexcitation mode k. By solving in first approximation the set of dynamical equations we obtain the (SE) decay rate for an atom placed at a distance z from one of the mirrors and with dipole moment μ orthogonal and parallel to the mirrors, respectively:

$$\Gamma_\perp(z) = A \times 3/(1-R)^2 \times (I_1 - I_2), \tag{2a}$$

$$\Gamma_\parallel(z) = A \times 3/2(1-R)^2 \times (I_1 + I_2), \tag{2b}$$

and

$$I_1(z) = \int_0^{\pi/2} G(\vartheta,z)\, d\vartheta,$$

$$I_2(z) = \int_0^{\pi/2} G(\vartheta,z)\cos^2(\vartheta)\, d\vartheta,$$

where A is the free-space rate, ϑ is the angle made by $\mathbf{k}$ with the cavity axis, $\beta=z/d$, $\delta=kd\cos(\vartheta)$, $E_k^0=a_k(0)\times\exp(-i\omega_k t)+\text{H.c.}$, $f_k=2\sin(\beta\delta)$, $R^2=R_1R_2$, R_1,R_2 = mirror reflectivities, ω = atomic resonant frequency, ϵ = field polarization, $g_k=-(2\pi\omega_k Y/n\hbar V)^{1/2}(\epsilon\cdot\mu)$, $Y=1/(1-R)^2[1+F\sin^2(\delta)]$ (Airy function), $F=4R/(1-R)^2=[(2/\pi)\times(\text{instrumental finesse})]^2$,

$$\gamma_k=2\pi(g_k f_k)^2[\delta(\omega_k-\omega)+\delta(\omega_k+\omega)],$$

$$G(\vartheta,z)=\sin^2(\beta\delta)\sin(\vartheta)/[1+F\sin^2(\delta)],$$

$k=2n\pi/\lambda$, V = volume of field definition. Equation (1a), expressing energy conservation, shows that the time evolution of the atomic energy operator depends on the full set of k modes available to the atom for spontaneous decay. Then the atomic decay rate Γ may be substantially affected by the cavity resonance only if all (or a large portion of the) k modes are confined by the cavity itself. Furthermore, by an averaging over the vacuum state, Eq. (1b) shows that the (SE) decay time T of the radiated energy $\langle 0|a_h^\dagger a_h|0\rangle$ detected under pulse excitation coincides with the decay time of the molecular excitation, i.e., $T=1/\Gamma$. On the other hand, Eq. (1b) also shows that if a CW field-intensity measurement is made on the observational mode h, the cavity transfer function Y plays a large role in the determination of the resonant behavior of the detected intensity, even in absence of any sizable resonant change of Γ, thus realizing in this last case a mere spatial-intensity-redistribution effect.

The experimental evidence of the variation of the spontaneous-emission lifetime T upon cavity tuning is reported in Fig. 2. Trace 2(a), representing (SE) "enhancement," is obtained for $d=\lambda/2$, while the SE "inhibition" effect, shown by trace 2(b), corresponds to $d=\lambda/8$. These data were taken with the periodic-optical-pumping angle set at $\vartheta_p=48°$. A 20% decrease of the effect was detected by our setting ϑ_p close to its extreme values 0° or 90°. At $\lambda=6328$ Å, we found that a lifetime shortening of a factor of at least 6, with respect to the free-space value T_0, could be easily detected, while the maximum time lengthening factor that we could obtain was $\simeq 3$. In addition, the pulse area was found to decrease by a factor of about 10 in passing from the maximum enhancement condition at $d=\lambda/2$ to the maximum inhibition that we could achieve. The theory expressed by Eqs. (1) and (2) shows that, in idealized conditions, nothing would prevent T from going to infinity for d approaching zero, and that the pulse area should remain constant for different values of d, apart from a small change due to volume variation. We explain this behavior by considering that, in our broad-band fluorescing system (band centered at $\bar{\lambda}=5950$ Å, $\Delta\bar{\lambda}=620$ Å), a sizable effect of excitation loss takes place as a result of cavity-resonance–assisted leaking of fluorescence photons mainly in cavity inhibition conditions: Molecular states (SE) inhibited at λ tend to deexcite in a Casimir cavity at $\lambda'<\lambda$ if a sizable effect of homogeneous broadening is present.[13]

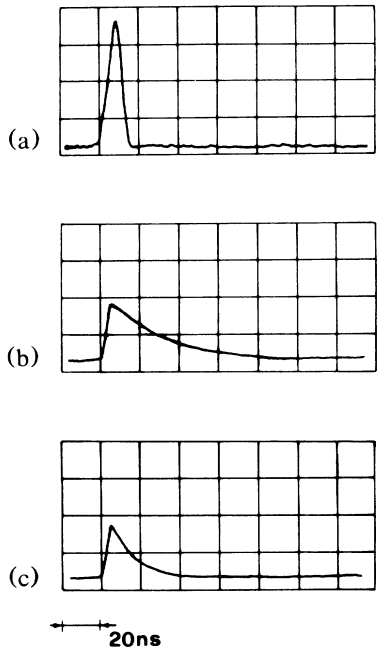

FIG. 2. Oscilloscope traces showing the Purcell effect in conditions of (a) spontaneous emission "enhancement" and (b) "inhibition" at $\lambda=6328$ Å. Traces (a) and (b) may be compared with (c) which shows the corresponding spontaneous emission decay in free space, i.e., in the absence of cavity effects.

In summary, we have given a most direct demonstration that the fundamental time of a quantum phenomenon, the spontaneous-emission time, is altered by a process of nonlocal space symmetry breaking, thus realizing a new type of Casimir effect.

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[a]On leave from the Physics Department, Universidade Estadual de Campinas, Brazil.

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[5]A preliminary account of the present work was presented at the International Conference Quantum Optics, Hamilton, New Zealand, 1986: F. De Martini and G. Innocenti, in *Quantum Optics IV,* edited by J. D. Harvey and F. D. Walls (Springer-Verlag, Berlin, 1986). That work reports the application of the microcavity to the study of SE.

[6]Light-intensity enhancement effects due to single-mirror interference have been reported by K. H. Drexhage, in *Progress in Optics,* edited by E. Wolf (North-Holland, New York, 1974), Vol. 12, p. 165, and by F. De Martini, Phys. Lett. A **115**, 421 (1986) and De Martini and Innocenti (Ref. 5). We should note that when the atoms are placed within a distance $z \lesssim \lambda$ from the single mirror, as in Drexhage's experiments, a substantial mode-confinement effect is realized (Ref. 5). Standing-wave effects due to refractive-index changes at the boundaries of fatty-acid layers deposited over one mirror are considered by Drexhage, *op cit.*, p. 178.

[7]In particular cases, the method based on frequency dependence of SE intensity can also be a reliable test: for instance, in a single-mode cavity, as for Jhe *et al.* (Ref. 3), or when an homogeneous-broadened SE line is present.

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[11]We note that in a plane Fabry-Perot microcavity additional allowed radiation modes are present. They correspond to waveguide-type field propagation in a direction parallel to the mirrors with polarization orthogonal to them. These modes are virtually unaffected by cavity resonance and their excitation has been found to be very weak in our experimental conditions.

[12]R. Loudon, *The Quantum Theory of Light* (Clarendon, Oxford, 1983), Chap. 5. Equations (1) are found to be independent of the operator-ordering procedure adopted in the nondifferential part of the Heisenberg equation. Cf. the "ordering"-problem works by Milonni, Ref. 2, and Dalibard *et al.*, Ref. 2. Note that when one coupling radiation mode is involved, as in our experiment, the Heisenbeg dynamical equations may be solved in closed form.

[13]The molecular fluorescence taking place in our work over a broad-band continuum of SE wavelengths suggests a further nice comparison with the Casimir effect where an analogous continuum of radiation modes is selected by the cavity. Note also in this connection that the SE "inhibition" condition tested in our experiment at $\lambda = 6328$ Å, actually extends over the entire set of fluorescence wavelengths $\lambda'' > \lambda$, thus giving rise to a (metastable) state of statistical nonequilibrium in the medium. Possibly, this peculiar condition is intrinsically interesting and deserves further investigation.