Anomalous Spontaneous–Stimulated-Decay Phase Transition and Zero-Threshold Laser Action in a Microscopic Cavity

F. De Martini and G. R. Jacobovitz[a]
Dipartimento di Fisica, Università di Roma 1, Roma, 00185 Italy
(Received 31 August 1987)

In an active "microcavity" the condition of maximum "enhancement" of spontaneous emission corresponds to resonant coupling of atoms with a single field mode. We demonstrate that this exceptional condition results in the effective cancellation of spontaneous emission, in the onset of stimulated emission at exceedingly low excitation levels, and in an anomalously high stimulated-emission gain. In the context of phase-transition theory the active microcavity behaves as a statistical ensemble undergoing an order-disorder transition at an extremely high value of the critical temperature.

PACS numbers: 42.50.Fx, 03.65.−w, 12.20.Fv, 32.50.+d

In recent papers we have reported the first realization of the optical "microscopic cavity" (or "microcavity"), i.e., a cavity with relevant dimension $d \leq \lambda$, the wavelength of the cavity-confined radiation.[1,2] With this apparatus the spontaneous-emission (SE) "inhibition-enhancement" process in atomic and molecular physics has been first demonstrated at optical frequencies.[2,3]

In the present paper we report the demonstration that the process of SE "enhancement" at $d = \bar{d} \equiv (\lambda/2)$ is effectively *nonexistent* in a microcavity as SE is overcome and merges into stimulated emission (StE) at exceedingly low levels of molecular excitation, thus realizing for the first time a virtually "zero-threshold-laser" action. This is a direct consequence of the properties of the microcavity at $d = \bar{d}$ consisting of a resonant single-mode field-atom interaction. This mode corresponds to a plane standing wave with **k** vector parallel to the Fabry-Perot (FP) cavity axis and with angular divergence $\theta \simeq f^{-1/2}$, where the "cavity finesse" is $f \gg 1$.[4,5]

Assume that under pulsed (or cw) excitation an atom or molecule with a homogeneous-broadened resonance at λ and linewidth $\Delta\lambda = \lambda\Delta\nu/\nu$, placed in a microcavity, emits a fluorescence photon at time $t = t_0$, and suppose that this is the only photon present in the cavity at that time. Formally, if $\hat{m}_k = a_k^\dagger a_k$ is the photon number operator for mode k, the quantum analysis applied to the atom-field interaction expresses the growth rate of the eigenvalue of $\hat{m}_k$ in the form $dm_k/dt = G^*(1+m_k)$. The emission of the first photon at $t = t_0$ spoils, for a time δt, the "vacuum state" corresponding to $m_k = 0$. Since no modes other than k are available, any other photon emitted in the cavity within δt is emitted over an "occupied state." Then it is not a SE photon in the usual sense, and its appearance determines the phase transition to the StE regime, i.e., to "order." For $m_k \gg 1$, the equation implies an exponential growth, i.e., the well-known condition of "StE gain."

Looking at this process in another perspective, we can say that the reduction of the dimensionality of the mode-statistical ensemble to a single degree of freedom, and the consequent elimination of the statistical-mode "reservoir" in the microcavity, lead to the establishment of a collective state in the medium if more than *one* atom, out of the large number N of excited ones, undergoes a radiative decay within δt. In this case, a long-range atom-atom correlation takes place (if appropriate conditions are met, by a "superradiant" process).[6,7] Of course, the transformation of *any* SE decay into a collective StE process, and then the absence of any fluorescence loss, leads to an anomalously large value of the StE gain. Thus, the only way to achieve the SE process (i.e., photon emission over the vacuum state, $|0\rangle$) in the presence of any low degree of excitation is to provide for the presence of only one atom in the microcavity, i.e., associating it with an atomic "trap."

The coherent atom-field interaction theory may be cast in a simple form.[2] Let H_k be the coupled atom-field Dicke Hamiltonian written in terms of the time-dependent atomic displacement operators $(\pi_i^\dagger, \pi_i)$ and field operators relative to the N atoms and the k mode, respectively. The Heisenberg equations written in terms of these operators, the "population-inversion" operator $\hat{\sigma} = (1/N)\sum_i [\pi_i^\dagger \pi^i - (\frac{1}{2})]$ and the average number of photons emitted over k, $\langle \hat{m}_k(t)\rangle = \langle m_k | a_k^\dagger a_k | m_k \rangle$, are (in the rotating-wave approximation)

$$-d(a_k^\dagger a_k)/dt = N\, d\hat{\sigma}/dt,$$
$$-d\langle \hat{m}_k\rangle/dt = \sum_i (g_k f_{ki}) \langle m_k | (\pi_i^\dagger E_k^0 + E_k^{0\dagger}\pi_i) | m_k \rangle - 2\sum_i \gamma_{ki} \langle m_k | (\pi_i^\dagger \pi_i) | m_k \rangle, \qquad (1)$$

where g_k and f_{ki} are the coupling constants expressing cavity-resonance effects and E_k expresses the initial conditions for the field operator.[8] In the single-mode case, Eqs. (1) can be solved in closed form (see Ref. 2). In particular, they lead to the expression of the absolute gain at $d=\bar{d}$: $\bar{g}=[3\pi k^3\sqrt{F}\,|\boldsymbol{\mu}|^2\bar{N}/2\hbar\omega]\propto G\bar{d}/c$, where $\boldsymbol{\mu}$ is the (randomly oriented) transition dipole moment.

For increasing $d>\bar{d}$, i.e., when additional "free" and "cavity-confined" modes become available for atomic deexcitation, the original orderly system at $d=\bar{d}$ becomes a "chaotic" system with rapidly increasing complexity. This leads to the appearance of mode competition and fluorescence loss with reduction of gain and an increase of the threshold-pumping level. The dynamics of the complex system is generally investigated by the methods of nonequilibrium statistical mechanics, i.e., by the Fokker-Planck method[9] and by second-order phase-transition theory.[10,11]

In the context of the latter theory, the behavior of the active microcavity may be understood by analogy with the phenomenologies of ferromagnetism and superconductivity. In our cooperative system the "ordering" process is so overwhelmingly "disorder" (here provided by cavity losses) that, once one photon is stored in the cavity, any additional single SE process provides the symmetry-breaking field to establish a phase transition to the state of nonzero average field $\langle E\rangle$, i.e., to the Glauber coherent-field state, $|\alpha\rangle$.[12] In nonequilibrium laser phase-transition theory the resulting virtual cancellation of the "disorder" phase leads to a zero value of the critical "reservoir variable" which is the "threshold population inversion," $\sigma_t\equiv\langle\hat{\sigma}\rangle_t=0$.[10] From this viewpoint, by analogy with equilibrium problems, our optical system may be thought of as behaving as an *extremely* high-critical-temperature (T_c) ferromagnet or superconductor.[10,11]

In our experiment a piezoelectrically tuned Fabry-Perot microcavity was formed by plane dielectric-coated mirrors, with $f=30$. A flow of sulforhodamine-640 (0.001 ethanol solution, free-space $T=3$ nsec) was kept between the mirrors. The optical pumping of the active medium was provided by second-harmonic generation at $\lambda_p=0.53$ μm by a self-injected Nd-doped yttrium aluminum garnet laser with $\simeq1.5$-nsec pulse duration.[13] The well-collimated laser beam was injected into the cavity through one mirror (A) with a selectable angle θ_p, taken with respect to the cavity axis, **z**, in order to take advantage of the effect of "periodic optical pumping," a technique allowing us by changes of θ_p to position the pumping zones and then to control the location of the excited molecules in the intracavity space.[1,14] $\theta_p=48°$ was found to correspond to a selective pumping located at $z=\bar{d}/2$ from the mirrors, i.e., in the central transverse section of the cavity. Mirror B, transmitting the detected fluorescence radiation, was 98% reflection coated for λ_p and λ, while mirror A was 99% reflection coated at λ and antireflection coated at λ_p. The pumping beam had a Gaussian intensity profile with mean diameter $=3$ mm. The fluorescence signal emitted from the cavity was filtered by an ir filter centered at $\lambda=6328$ Å ($\delta=1$ Å passband), focused by a dioptric system with focal length $f'=180$ mm on the 4-mm-diam end of an optical fiber, and then recorded by an RCA model C31034A photomultiplier (rise time <1.5 nsec). The cavity alignment was maintained by means of a He-Ne laser through two disk-shaped PZT transducers, while the tuning at $d=\bar{d}$ was controlled by a Micro-Controle positioner by 1000-Å steps and, on a finer scale, by a large toroidal (PZT) transducer. The signal wave forms were recorded with a LeCroy model 8013A fast digitizer while the intensities of the signal and pump beams were recorded through a computer-interfaced analog-to-digital system.

At the SE-enhancement condition, $d=\bar{d}$, the signal pulse width was found to coincide with that of the "pump" pulses at any lowest molecular-excitation level, as expected for StE. The onset of the StE exponential-gain regime (the laser phase transition) taking place at the anomalous threshold pumping intensity, $I_p\propto\sigma_t=0$ for $d=\bar{d}$, is shown in Fig. 1. In Fig. 1, the analogous transition for $d=5\lambda$ is reported for comparison. At $d=\bar{d}$ we were not able to determine experimentally a lower limit of the signal intensity for the transition (i.e., we could not find any departure from the exponential law) in spite of the fact that our experimental conditions were not the the idealized ones considered at the beginning of this paper. In fact, a real zero-threshold laser transition should have been found with our technique

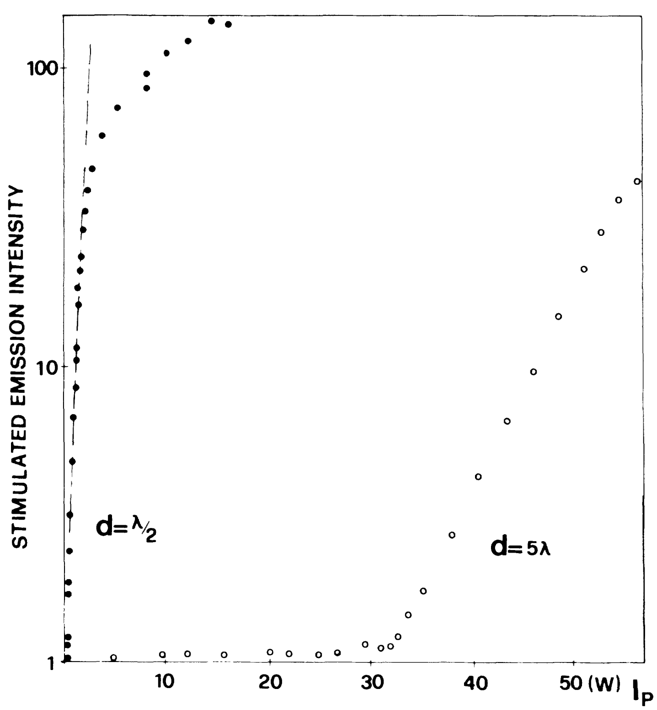

FIG. 1. Laser phase transitions for microcavity dimensions $d=\bar{d}\equiv\lambda/2$ and $d=5\lambda$. The emitted intensities shown for $d=5\lambda$ should be multiplied by 10 to be compared with the $\bar{d}$ data.

only if the cavity radial modes were absent[5] and if the linewidth of the detected radiation had been $\Delta\nu = 1/T_0 = 1/t_c$ and $t_c < \tau$, the output StE pulse length. In our case, however, because of the inhomogeneously broadened detection bandwidth, t_c must be evaluated by the replacement of $\Delta\nu$ with $\delta\nu/\lambda \gg \Delta\nu$. By consideration of the cavity-Q enhancement effect, the number of detected StE photons at threshold for $d = \bar{d}$ is calculated to be $\simeq 30$, which corresponds approximately to the limit sensitivity of our apparatus. The pump intensity I_p reported in Fig. 1 has been evaluated to correspond to the amount of excitation released to the active medium over δ. The slope of the exponential-gain sections, below the curve-bending saturation regions, in the plots of Fig. 1, is proportional to the StE gain. Note that for $d = 5\lambda$ the gain is *smaller* than for $d = \bar{d}$, in spite of the tenfold increase in the active population. This is but one consequence of the privileged dynamical condition at $d = \bar{d}$, as we already stressed, and is associated with the appearance at $d > \bar{d}$ of additional transverse modes as a result of the explicit form of $Y(\theta, d)$ and to the increasingly imperfect cavity mode confinement, for increasing d. As already stated, the set of modes appearing for $d > \bar{d}$ coincides with the statistical mode reservoir which is responsible for the phase transition at $(I_p)_t \propto \sigma_t > 0$, as shown in Fig. 1 for $d = 5\lambda$. The large efficiency of the StE process at $d = \bar{d}$ is indicated by the value of the relative gain evaluated on the basis of Eqs. (1): $g = 1.5 \times 10^5$ cm/kW, a result we found to be in order-of-magnitude agreement with the experimental data.

Note that the strikingly anomalous behavior of atoms when confined in an active microcavity is *not* obtainable by the d-dimensional scaling of the usual active macroscopic-cavity effects. On the contrary, it is in fact a quantum effect reminiscent of the nonlocal space-symmetry–breaking confinement processes taking place over the scale of the de Broglie wavelength of the test particle, λ. Examples of such effects are the electromagnetic Casimir effect,[15] all kinds of particle diffraction, and the electron-energy quantization in semiconductor multiple quantum wells. This kind of behavior is of course quite general.

The above results, which arise from an extreme "vacuum-confinement" process, can be generalized to all physical situations in which the vacuum field is at the origin of a "spontaneous" emission process as, for instance, within the phenomenologies of stimulated (Raman or Brillouin) scattering and of "optical parametric oscillation."[10] The "zero-threshold" effect, introduced in the present Letter, is expected to find relevant applications in the field of solid-state optoelectronics as for instance in connection with the microminiaturization of active optical elements in integrated semiconductor structures or with coherent-light generation in active single-mode optical fibers.

We acknowledge the assistance of P. Mataloni and stimulating discussions with M. O. Scully and P. W. Milonni. The work of one of us (G.R.J.) is supported by the International Center for Theoretical Physics, Trieste, Italy.

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

[1] F. De Martini and G. Innocenti, in *Quantum Optics IV*, edited by J. D. Harvey and D. F. Walls (Springer-Verlag, Berlin, 1986).

[2] F. De Martini, G. Innocenti, G. R. Jacobovitz, and P. Mataloni, Phys. Rev. Lett. **59**, 2955 (1987); W. Jhe, A. Anderson, E. A. Hinds, D. Meschede, L. Moi, and S. Haroche, Phys. Rev. Lett. **58**, 666 (1987).

[3] E. M. Purcell, Phys. Rev. **69**, 681 (1946).

[4] M. Born and E. Wolf, *Principles of Optics* (Macmillan, New Yor, 1964), Chap. 7.

[5] The electromagnetic mode density and the angular distribution of the radiation emitted by an active microcavity are expressed as functions of d and θ by the cavity transfer function appearing in the context of the theory of the passive Fabry-Perot interferometer. This function is the Airy function $Y(\theta,d) = 1/(1-R)^2(1+F\sin^2\delta)$, where $R^2 = R_1 R_2$, $R_{1,2}$ = mirror reflectivities, $F = (2f/\pi)^2$; $\delta = kd\cos\theta$, $k = 2\pi n/\lambda$, n = refractive index. Strictly speaking, the microcavity single-mode concept is an approximate one. In fact, in a Fabry-Perot cavity waveguide-type "radial" modes propagating in directions parallel to the mirrors with polarization orthogonal to them are also allowed; however, they are unaffected by cavity resonance and have been found to be weakly excited for $d = \bar{d}$.

[6] W. Heitler, *The Quantum Theory of Radiation* (Clarendon, Oxford, 1960), p. 60; R. Loudon, *The Quantum Theory of Light* (Clarendon, Oxford, 1983), Chap. 4; R. H. Dicke, Phys. Rev. **93**, 99 (1954).

[7] For superradiant-type coherence processes in maser work, cf. P. Goy, M. Raimond, M. Gross, and S. Haroche, Phys. Rev. Lett. **50**, 1903 (1983); R. G. Hulet, E. S. Hilfer, and D. Kleppner, Phys. Rev. Lett. **55**, 2137 (1985); D. Meschede, H. Walther, and G. Müller, Phys. Rev. Lett. **54**, 551 (1985); P. Filipovicz, P. Meystre, G. Rempe, and H. Walther, Opt. Acta **32**, 1105 (1985).

[8] In Eq. (1), $g_k = -(2\pi\omega_k Y/n\hbar V)^{1/2}\epsilon\cdot\boldsymbol{\mu}$; V = field-definition volume; ϵ = field polarization; $f_{ki} = 2\sin(\beta\delta)$; $\beta = z/d$; θ = angle made by $\mathbf{k}$ with the cavity axis; $\gamma_{ki} = 2\pi(g_k f_{ki})^2[\delta(\omega_k - \omega) + \delta(\omega_k + \omega)]$; $E_k^0 = a_k(0)\exp(-i\omega_k t) + \text{H.c.}$; ω = atomic resonance frequency. The ket $|m_k\rangle$ appearing in Eqs. (1) represents the state vector defined in the Hilbert space of the atom-field system.

[9] W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973), Chaps. 6 and 8.

[10] M. Sargent, III, M. O. Scully, and W. E. Lamb, *Laser Physics* (Addision-Wesley, Reading, MA, 1974), Chap. 16; V. De Giorgio and M. O. Scully, Phys. Rev. A **2**, 1170 (1970); J. F. Scott, M. Sargent, III, and C. Cantrell, Opt. Commun. **15**, 13 (1975).

[11] Notice that in the passage from nonequilibrium to equilibrium problems, there is a sign reversal in the inequalities involving reservoir variables and corresponding to the same "phase." Then the "zero-threshold laser" condition $\sigma_t = 0$ implying

nonexistence of the "incoherent-field phase" corresponds in the analogy to an infinite value of the equilibrium "order parameter," T_c. The concept of "negative temperature" in early maser theory is related to this analogy.

[12]R. Glauber, Phys. Rev. **130**, 2529 (1963); C. Quigg, *Gauge Theories of the Strong, Weak and Electromagnetic Interactions* (Benjamin, London, 1983), Chap. 5, reports a modern presentation of the symmetry-breaking concept in the context of gauge theories.

[13]C. H. Brito Cruz, E. Palange, and F. De Martini, Opt. Commun. **39**, 331 (1981); S. Giacomini, G. Innocenti, P. Mataloni, and F. De Martini, Appl. Opt. **26**, 3179 (1987).

[14]F. De Martini, Phys. Lett. A **115**, 421 (1986); A. Kastler, Appl. Opt. **1**, 17 (1962).

[15]H. B. G. Casimir, Proc. Ned. Akad. Wet. **60**, 793 (1948); C. Itzykson and J. B. Zuber, *Quantum Field Theory* (McGraw Hill, New York, 1980), Chap. 3; G. Plunien, B. Muller, and W. Greiner, Phys. Rep. **134**, 89 (1986).