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CAVITY QUANTUM ELECTRODYNAMICS

A new generation of experiments shows that spontaneous radiation from excited atoms can be greatly suppressed or enhanced by placing the atoms between mirrors or in cavities.

Serge Haroche and Daniel Kleppner

Ever since Einstein demonstrated that spontaneous emission must occur if matter and radiation are to achieve thermal equilibrium, physicists have generally believed that excited atoms inevitably radiate.[1] Spontaneous emission is so fundamental that it is usually regarded as an inherent property of matter. This view, however, overlooks the fact that spontaneous emission is not a property of an isolated atom but of an atom–vacuum system. The most distinctive feature of such emission, irreversibility, comes about because an infinity of vacuum states is available to the radiated photon. If these states are modified—for instance, by placing the excited atom between mirrors or in a cavity—spontaneous emission can be greatly inhibited or enhanced.

Recently developed atomic and optical techniques have made it possible to control and manipulate spontaneous emission (figure 1). Experiments have demonstrated that spontaneous emission can be virtually eliminated or else made to display features of reversibility: Instead of radiatively decaying to a lower energy state, an atom can exchange energy periodically with a cavity.

Serge Haroche is a professor of physics at the University of Paris VI and at the Ecole Normale Supérieure, in Paris, and at Yale University, in New Haven, Connecticut. **Daniel Kleppner** is Lester Wolfe Professor of Physics at the Massachusetts Institute of Technology, in Cambridge, Massachusetts.

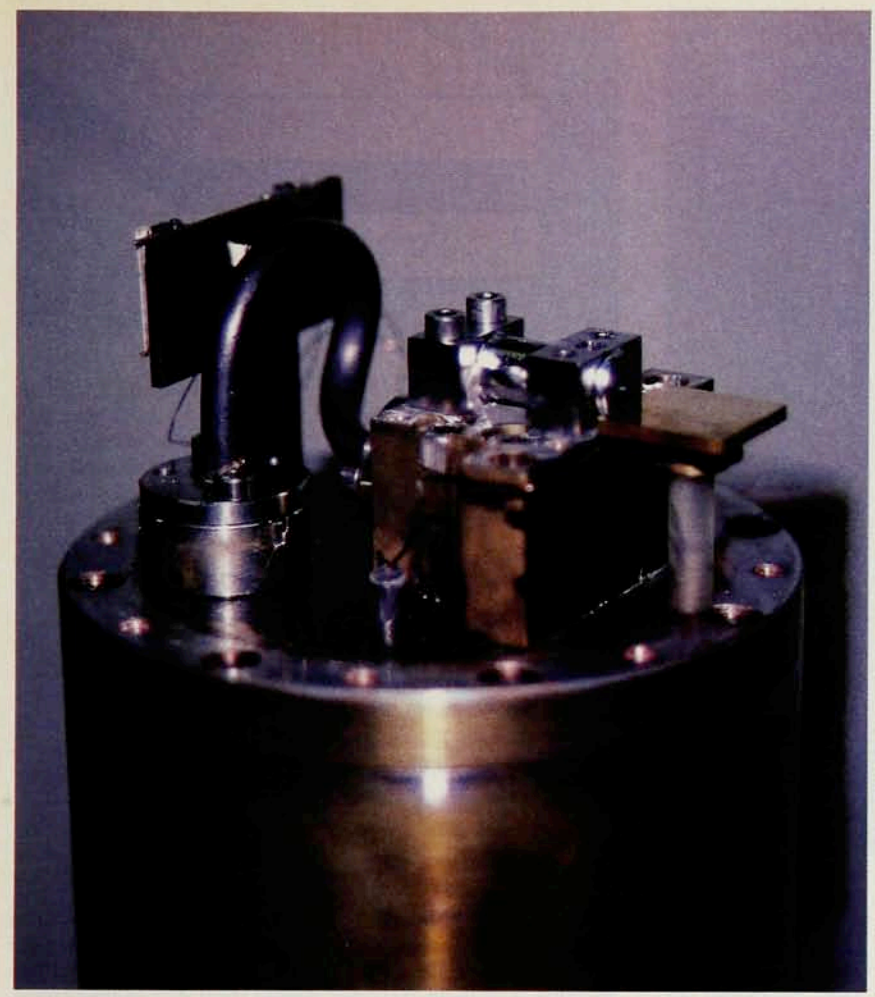
The recent research on atom–vacuum interactions belongs to a new field of atomic physics and quantum optics called cavity quantum electrodynamics. In addition to demonstrating dramatic changes in spontaneous emission, cavity QED has led to the creation of new kinds of microscopic masers that operate with a single atom and a few photons or with photons emitted in pairs in a two-photon transition.

Emission in free space

We can introduce cavity QED with a brief review of spontaneous emission in free space. Consider a one-electron atom with two electronic levels e and f separated by an energy interval $E_e - E_f = \hbar\omega$. Spontaneous emission appears as a jump of the electron from level e to level f accompanied by the emission of a photon. This process can be understood as resulting from the coupling of the atomic electron to the electromagnetic field in its "vacuum" state.

A radiation field in space is usually described in terms of an infinite set of harmonic oscillators, one for each mode of radiation. The levels of this oscillator correspond to states with 0, 1, 2, . . . , n photons of energy $\hbar\omega$. In its ground state each oscillator has a "zero-point" energy $\hbar\omega/2$ associated with its quantum fluctuations.

The rms vacuum electric-field amplitude E_{vac} in a mode of frequency ω is $[\hbar\omega/(2\epsilon_0 V)]^{1/2}$, where ϵ_0 is the permittivity of free space, V is the size of an arbitrary quantization volume and the units are SI. The coupling of the atom to each field mode is described by the elementary



Resonant superconducting cavity. The cylindrical niobium cavity, situated between the two shiny, rectangular blocks at the top of the apparatus, is used for cavity QED experiments on Rydberg atoms at the Ecole Normale Supérieure in Paris. It is cooled by a helium cryostat, the large cylinder at the bottom of the photograph. The atomic beam passes along the cavity axis, entering and leaving through small holes. **Figure 1**

frequency

$$\Omega_{ef} = D_{ef}E_{vac}/\hbar$$

Here D_{ef} is the matrix element of the electric dipole of the atom between the two levels, and Ω_{ef}, which is often referred to as the Rabi frequency of the vacuum, is the frequency at which the atom and the field would exchange energy if there were only a single mode of the field. An essential feature of spontaneous emission in free space, however, is that the atom can radiate into any mode that satisfies the conservation of energy and momentum. The time of emission and the particular mode in which the photon is observed are random variables.

The probability Γ_0 of photon emission per unit time, more familiarly called the Einstein A-coefficient, is proportional to the square of the frequency Ω_{ef} and to the mode density $\rho_0(\omega)$, the number of modes available per unit frequency interval. The mode density is given by the expression $\rho_0(\omega) = \omega^2 V/\pi^2 c^3$, where it is assumed that the quantization volume V is large compared to λ^3, or $(2\pi c/\omega)^3$. The probability Γ_0 is given by the Fermi "golden rule":

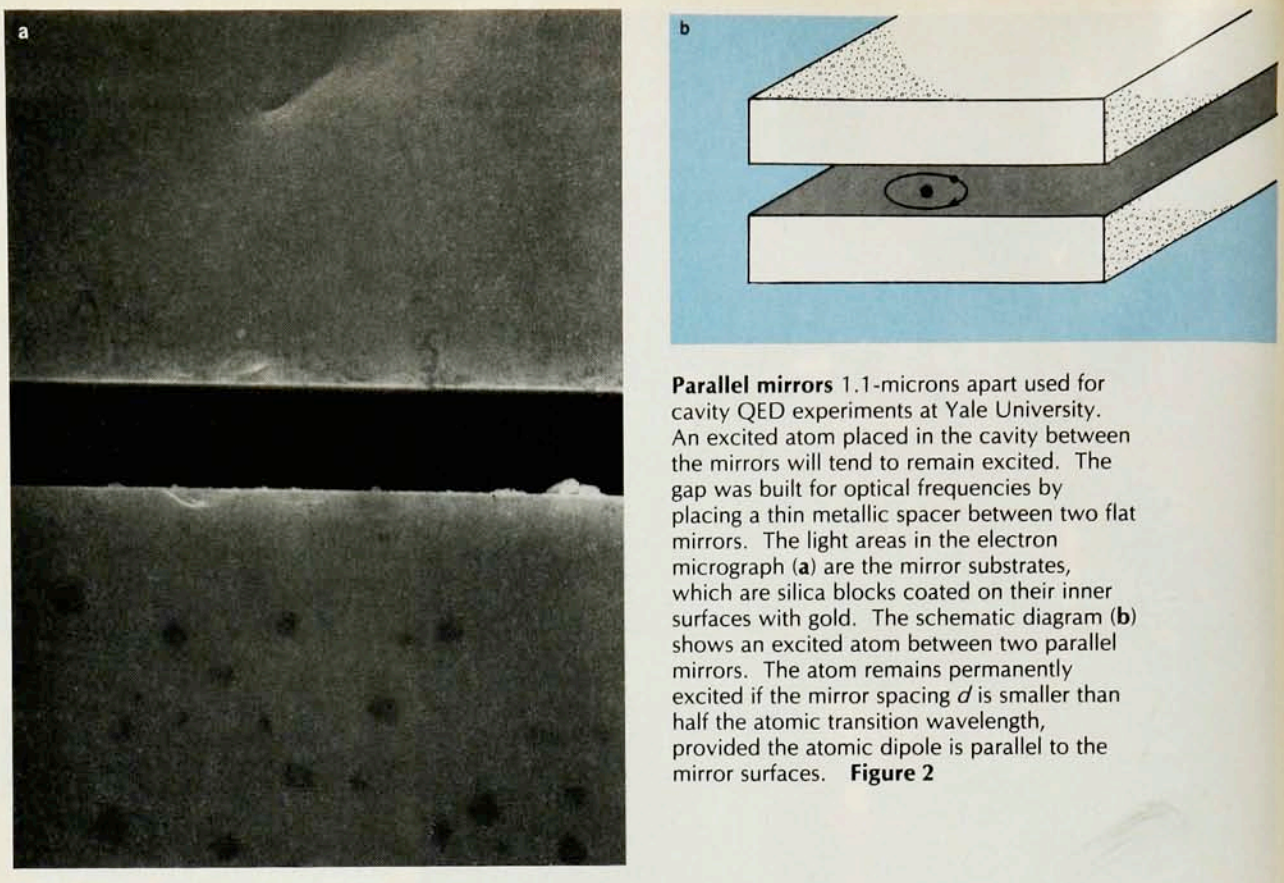
$$\Gamma_0 = 2\pi\Omega_{ef}^2 \frac{\rho_0(\omega)}{3} = \frac{\omega^3}{3\pi\hbar c^3} \frac{|D_{ef}|^2}{\epsilon_0}$$

The probability $P_e(t)$ of finding an atom still excited at time t after its preparation in state e is $\exp(-\Gamma_0 t)$. Such an exponential decay law describes an irreversible process that leads the atom irrevocably to its ground state. The source of irreversibility is the continuum of field modes resonantly coupled to the atom. The vacuum field acts as a gigantic "reservoir" in which the atomic excitation decays away.

Emission between mirrors

The mode structure of the vacuum field is dramatically altered in a cavity whose size is comparable to the wavelength. The schematic diagram in figure 2 shows an atom confined between two plane, parallel mirrors with separation d. For an electric field polarized parallel to the mirrors no mode exists unless $\lambda < 2d$: As λ is increased, the lowest-order mode cuts off abruptly when λ exceeds $2d$. As a result, an excited atom whose radiation arises from an electric dipole moment oscillating parallel to the mirrors becomes infinitely long-lived when $\lambda > 2d$. Classically, the electron orbit of such an atom is essentially parallel to the mirrors; we call this σ polarization. By contrast, for polarization normal to the mirrors—π polarization—there is no such cutoff and the excited state decays rapidly. Such simple cavity structures are easy to build in the microwave domain and can now be realized down to the micron size for optical fields, as the photograph in figure 2 shows.

The influence of boundary conditions on atomic radiation was pointed out long ago, and a considerable body of literature exists on the subject.[2] Until recently, however, experimental evidence for these effects had been scarce because of the difficulties of preparing, controlling



Parallel mirrors 1.1-microns apart used for cavity QED experiments at Yale University. An excited atom placed in the cavity between the mirrors will tend to remain excited. The gap was built for optical frequencies by placing a thin metallic spacer between two flat mirrors. The light areas in the electron micrograph (**a**) are the mirror substrates, which are silica blocks coated on their inner surfaces with gold. The schematic diagram (**b**) shows an excited atom between two parallel mirrors. The atom remains permanently excited if the mirror spacing d is smaller than half the atomic transition wavelength, provided the atomic dipole is parallel to the mirror surfaces. **Figure 2**

and observing the emission of isolated atoms or molecules near surfaces. In the early 1970s Karl H. Drexhage at the University of Marburg, Germany, carried out pioneering work on the fluorescence of organic dyes deposited on dielectric films over a metallic mirror.[3] He observed alterations in the rate and pattern of the emission, but the open geometry of his experimental setup prevented any major reduction of the spontaneous emission rate. More recently several groups have studied isolated atomic radiators in cavities, and have demonstrated the inhibition of spontaneous emission at wavelengths from the microwave to the optical. A group at the University of Washington performed the first in this generation of experiments, observing the radiation damping of an isolated electron undergoing cyclotron motion in an electromagnetic trap whose walls formed a high-mode cavity. They discovered large changes in the cyclotron damping rate that depended on the position of the electron with respect to the trap's nodal pattern.[4]

Physicists at MIT, Yale and the University of Rome have carried out experiments using the parallel-mirror geometry of figure 2. The MIT experiment demonstrated the inhibition of spontaneous emission from Rydberg states of cesium atoms in a beam.[5] The atoms radiated at a wavelength of about 0.4 mm as they passed between two 20-cm-long aluminum mirrors separated by approximately 0.2 mm. Just before entering the "cavity," the atoms were prepared in such a way that their radiating dipoles were strictly parallel to the mirrors, much like the "circular" state sketched in figure 2. The atoms surviving in the initial quantum state were detected at the cavity exit by ionizing them in a small electric field. Atoms that underwent spontaneous emission were transferred to a more tightly bound level that remained un-ionized.

Figure 3 shows how the signal of the atoms changes as the ratio $\lambda/2d$ is varied around the critical value of 1. The large increase in transmission when λ exceeds $2d$ is clear evidence of the inhibition of spontaneous emission. The lifetime is at least 20 times longer than it is in free space. The decrease for $\lambda/2d > 1.015$ is an artifact caused by the ionization of the atoms by the electric field.

The experimenters at Yale[6] suppressed spontaneous emission in the near infrared using a geometry similar to the MIT experiment but with the much smaller waveguide structure shown in figure 2a. The Yale experiment employed a cesium atomic beam, excited into the low-lying $5d$ level. The inhibited transition was $5d \rightarrow 6p$ at a wavelength of 3.5 microns, far beyond the cutoff wavelength of 2.2 microns. Excited atoms propagated through the tunnel for about 13 natural lifetimes without appreciable decay. Application of a small magnetic field to change the orientation of the atomic dipoles demonstrated the anisotropy of spontaneous emission between mirrors. If the magnetic field has a component parallel to the mirror surface, the dipole precesses so that it acquires a perpendicular component. After rotating through only a small fraction of a turn the dipole can spontaneously radiate a π-polarized photon. Figure 4 shows the transmission of excited atoms through the microtunnel as a function of the angle between the magnetic field and the normal to the mirrors: The large change in the lifetime induced by the magnetic precession is evident. In another experiment, carried out at the University of Rome, the radiative lifetime of dye molecules at optical frequencies was lengthened using a cavity formed by plane, parallel mirrors.[7]

Resonant cavity emission

Just as a cavity below cutoff suppresses vacuum fluctuations, a resonant cavity enhances them. How an atom

behaves in a resonant cavity depends on the ratio of the vacuum Rabi frequency to the cavity bandwidth, $\Omega_{ef}/\Delta\omega_c$. The cavity bandwidth is most conveniently described by the quality factor Q, which is given by $\omega/\Delta\omega_c$. The reciprocal of $\Delta\omega_c$ is effectively the density of modes "seen" by the atom in the cavity; alternatively, it is the lifetime of a photon in the cavity. In open structures such as the one shown in figure 2a, edge diffraction limits the quality factor Q. By employing spherical mirrors to create a Fabry–Perot resonator, Q can be enhanced substantially. In the microwave regime, closed cavities are practical (figure 1). A superconducting microwave cavity can achieve values of Q up to 10^{11} and photon storage times of a fraction of a second.

In a low-Q cavity the emitted photon is damped rapidly and an atom undergoes radiative decay much like it does in free space, though at an enhanced rate. The radiation rate in a cavity of volume V is

$$\Gamma_{cav} \cong \Gamma_0 \frac{Q\lambda^3}{V}$$

Compared with the rate in free space, the emission rate is increased by the ratio $Q\lambda^3/V$, which can be large. Physicists at École Normale Supérieure in Paris have observed this regime of enhanced spontaneous emission in the millimeter wave regime for Rydberg atoms of sodium coupled to a Fabry–Perot cavity.[8] The spontaneous emission rate was enhanced by a factor of approximately 500.

In the optical regime, physicists in the Rome group[7] and at the MIT Spectroscopy Laboratory[9] have observed effects of both enhanced and inhibited spontaneous emission. The MIT experimenters employed a spherical Fabry–Perot resonator. A laser within the resonator excited an atomic beam of ytterbium, which fluoresces at a wavelength of 556 nm. As the experimenters tuned the resonator through successive resonances, the radiation rate into the solid angle subtended by the resonator was alternately inhibited by a factor of 42 and enhanced by a factor of 19.

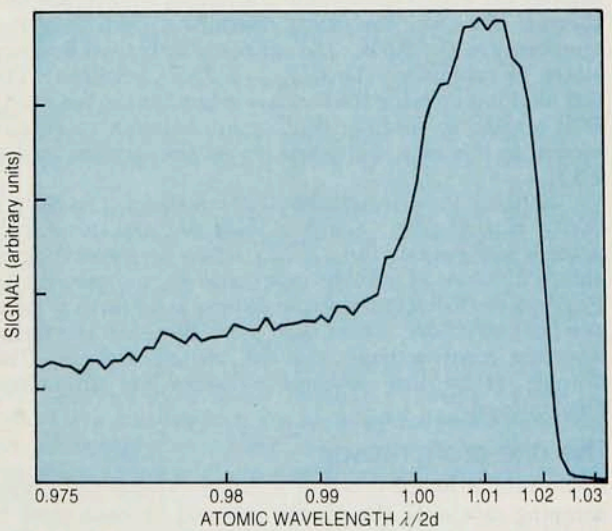
The regime of very high Q, where $\Omega_{ef}/\Delta\omega_c > 1$, manifests totally new behavior. The radiation remains in the cavity so long that there is a high probability it will be reabsorbed by the atom before it dissipates. Spontaneous emission becomes reversible as the atom and the field exchange excitation at the rate Ω_{ef}. Such behavior is a well-known feature of the interaction of an atom with a classical monochromatic field. These so called "Rabi oscillations" are familiar in nuclear magnetic resonance and optical transient experiments. In cavity QED, however, the atom couples to its own one-photon field without any externally applied radiation.

The link between classical Rabi oscillations and spontaneous Rabi oscillations is described by the equation below for the probability $P_e(t)$ of finding the atom in its excited state at time t, assuming it was prepared in this state at time 0. The probability that there are n photons in the cavity is described by the distribution function $p(n)$, and the cavity is taken to have an infinite quality factor Q.

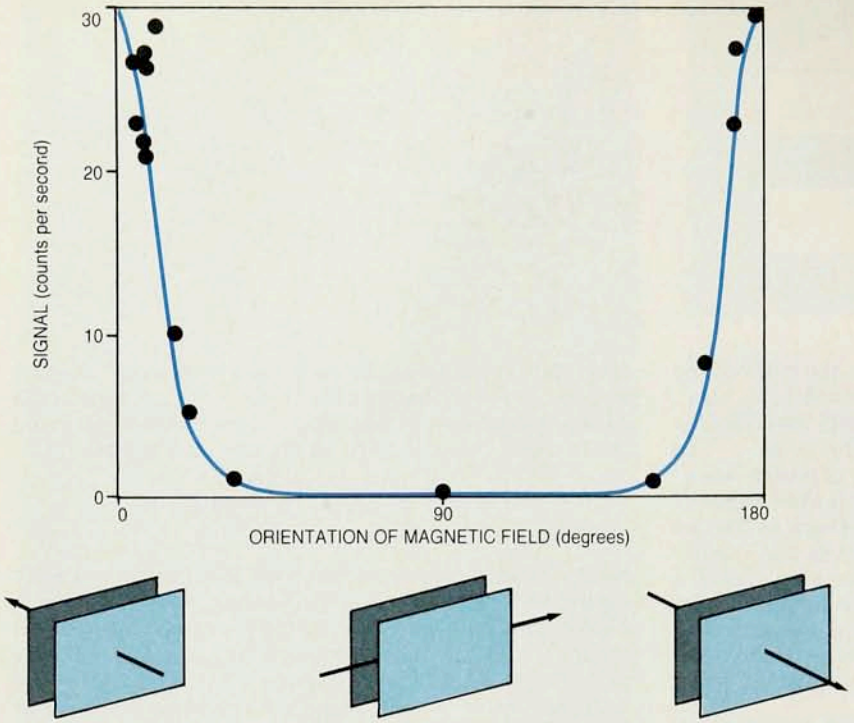
$$P_e(t) = \sum_n p(n) \cos^2(\tfrac{1}{2}\Omega_{ef}\sqrt{n+1}\, t)$$

Spontaneous emission corresponds to an initially empty cavity: $p(0) = 1$. A classical field corresponds to a narrow photon distribution peaked around a large average photon number $\bar{n}$. (The Rabi frequency $\Omega_{ef}\sqrt{n+1}$ is roughly proportional to the square root of n, that is, to the electric field amplitude in the mode.) For ordinary atoms or molecules, Ω_{ef} is intrinsically small, and observing the Rabi oscillation requires an enormous average photon number $\bar{n}$. For Rydberg states, on the other hand, Ω_{ef} can be relatively large—10^3 to 10^6 sec^{-1} for principal quantum numbers around 40—and the Rabi oscillations become observable even with $\bar{n} = 0$.

Physicists at the Max Planck Institute for Quantum Optics near Munich have observed Rabi oscillations induced by a small thermal field in the cavity.[10] The



Survival of excited Rydberg atoms moving in a gap between mirrors, plotted as a function of the wavelength for spontaneous emission in the vicinity of the cavity cutoff.[5] The signal comes from excited atoms detected at the cavity exit. The atomic wavelength λ is varied by applying a small electric field to the atoms. The sharp increase in survival when the ratio $\lambda/2d$ is 1 is caused by the inhibition of spontaneous emission. **Figure 3**



Transmission of excited atoms through a mirror gap causing a cutoff of spontaneous emission in the infrared regime around 3.5 microns.[6] The gap structure is shown in figure 2a. The signal is plotted against the angle between a small applied magnetic field and the normal to the mirrors. The diagrams at the bottom of the figure illustrate the magnetic field orientations defined as 0°, 90° and 180°. At zero angle the atoms are polarized parallel to the mirrors and the large transmission of excited atoms is evidence of inhibited spontaneous emission. When the angle is nonzero, Larmor precession reorients the atomic dipoles so that they can emit π-polarized radiation, which can propagate in the gap. As a result, the atoms radiate spontaneously and fail to reach the detector in their excited state. **Figure 4**

photon number distribution is then given by the Planck formula: $p(n) = [1 - \exp(-\hbar\omega/k_B T)] \exp(-n\hbar\omega/k_B T)$, where k_B is the Boltzmann constant. The pattern of oscillations is a complicated function of time resulting from the beating between the elementary Rabi frequencies weighted by the corresponding values of $p(n)$. Figure 5 shows the probability $P_e(t)$ as a function of the time of interaction with a cavity field containing an average of 2 thermal photons. The cavity operated at 21.6 GHz at a temperature T of 2.5 K. The experimenters used Rydberg states of rubidium—the $63P_{3/2} \rightarrow 61D_{5/2}$ transition. The flux of atoms crossing the cavity was low, and so the cavity field relaxed to thermal equilibrium between successive atoms. In this way, each atom probed the thermal field at 2.5 K.

Rabi oscillations induced by small thermal fields in a cavity can display "quantum collapse and revival."[11] After a few periods the various terms corresponding to different values of n in the expression for the probability $P_e(t)$ interfere destructively, and there is no further trace of a beat structure. Some time later, however, the terms interfere constructively and the beating revives. The Munich group has obtained evidence for this novel phenomenon.[10]

The one-atom maser

If the rate of atoms crossing a cavity exceeds the cavity damping rate ω/Q, the photon released by each atom is stored long enough to interact with the next atom. The atom–field coupling becomes stronger and stronger as the field builds up, eventually evolving into a steady state. The system is a new kind of maser, which operates with exceedingly small numbers of atoms and photons.[12] Atomic fluxes as small as 100 atoms per second have generated maser action. For such a low flux there is never more than a single atom in the resonator—in fact, most of the time the cavity is empty. These "micromaser" devices were operated for the first time at the Max Planck Institute for Quantum Optics.[13]

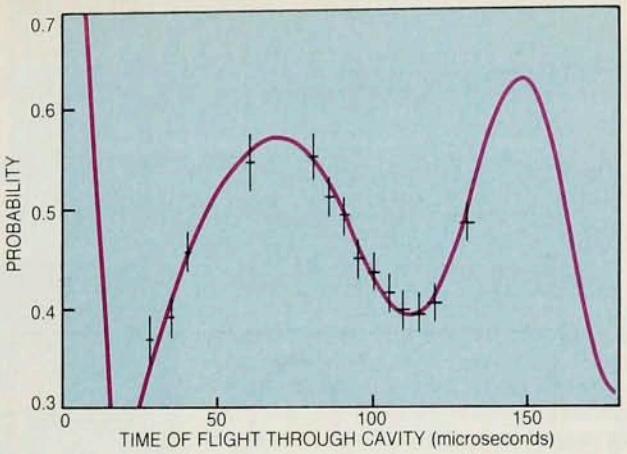
It is interesting to analyze how the field grows in an ideal micromaser operating at 0 K with a fixed atom-cavity interaction time t_{int}. Assume there is an ideal cavity (infinite Q) and an ideal field ionization detector that allows us to determine the state e or f in which each atom leaves the cavity. After the first atom (atom #1) has crossed the resonator, the state of the atom–field system is a linear superposition of states $|e,0\rangle$ and $|f,1\rangle$. (The notation indicates an atom in state e correlated with 0 photons in the cavity and an atom in state f correlated with one photon.) Using standard notation we can write

$$|\Psi_1\rangle = \cos(\Omega_{ef} t_{int}/2)\,|e,0\rangle + \sin(\Omega_{ef} t_{int}/2)\,|f,1\rangle$$

If $\Omega_{ef} t_{int}$ is small, 0.1 for example, there is a large probability of finding atom #1 in state e. If this occurs, then immediately thereafter $|\Psi_1\rangle = |e,0\rangle$, and so the field will contain exactly zero photons. The next atom will then interact with an empty cavity. At some time, however, a first atom, say atom N, will be found in state $|f\rangle$, correlated to the instantaneous "appearance" of one photon in the cavity. Atom $(N+1)$ will then undergo a different evolution: The state of the system "atom $(N+1)$ + cavity field" after atom $(N+1)$ has left the cavity will be

$$|\Psi_{N+1}\rangle = \cos(\Omega_{ef}\sqrt{2}\,t_{int}/2)\,|e,1\rangle + \sin(\Omega_{ef}\sqrt{2}\,t_{int}/2)\,|f,2\rangle$$

Continuous monitoring of the system would reveal a kind of random walk of the photon number: Each time an atom is detected in state $|f\rangle$ the field has gained one photon. The process is essentially random because the probability for an atom to flip from e to f is governed by quantum mechanical chance. It is a random process with *memory*, however, because the probability law for each step depends on the outcome of the previous steps. This simple build-up process has been studied theoretically and provides one of the simplest illustrations of the quantum theory of measurement.[14] The field can be viewed as a quantum harmonic oscillator. The two-level atoms cross-



Probability of finding an atom in the upper level of a two-state system as a function of the atom–cavity interaction time.[16] The data were taken in a cavity tuned to 21.6 GHz, the frequency of the $63P_{3/2} \rightleftarrows 61D_{5/2}$ transition of Rb^{85}. The curve is theoretical. Radiation is induced by the thermal field in the cavity at 2.5 K. The Rabi oscillation is clearly observable. **Figure 5**

ing the cavity play a double role: They modify the field by the quantum mechanical "kicks" they deliver, and they serve as measurement devices that detect the field as it grows. A steady state is reached if there exists a photon number n_0 such that the quantity $(\Omega_{ef}\sqrt{n_0+1}\ t_{int})/2$ is an integral multiple of π. The photon number remains trapped with the value n_0 because each subsequent atom will leave the cavity in the e state. Such a radiation state is highly nonclassical, for the field has a precisely defined energy but a completely random phase. It has no amplitude fluctuations, whereas an ordinary electromagnetic field has quantum-limited intensity fluctuations proportional to $\sqrt{\bar{n}}$. Such nonclassical states of microwave radiation fields are now being studied at the Max Planck Institute for Quantum Optics.

The two-photon maser.

At the Ecole Normale Supérieure physicists have created a microwave maser that uses a Rydberg atom–cavity system but that operates on a two-photon transition.[15] In two-photon emission an atom simultaneously radiates a pair of photons with frequencies ω_1 and ω_2 while jumping from an initial level e to a final level f. Energy conservation requires that $E_e - E_f = \hbar(\omega_1 + \omega_2)$. Because two-photon emission is a second-order process, it usually occurs only from metastable levels that are forbidden to radiate by a single-photon process because of some selection rule. If one- and two-photon emission channels are both open for the decay of an atomic level, the one-photon process dominates and two-photon effects are negligible. In the level configuration of figure 6a, which is often found in the structure of Rydberg atoms, the upper level e can decay by one-photon emission to the opposite parity level i or by two-photon emission to the same parity level f. In free space the former process is overwhelmingly more probable. In a cavity that is resonant at a frequency ω equal to $(E_e - E_f)/2\hbar$, however, the spontaneous emission of two "degenerate" photons at frequency ω is enhanced, whereas the one-photon rate is suppressed.

This effect has been experimentally demonstrated in a beam of rubidium atoms prepared in the 40S state. The atoms traversed a superconducting cavity tuned to 68.4 GHz, half the frequency of the 40S→39S transition. The resonant transfer of atoms between these two levels, shown in figure 6b, demonstrates two-photon maser action. This new quantum oscillator displays unique features because the photons are emitted in pairs. For instance, startup of the system is characterized by long delays, very different from the prompt startup of a one-photon maser.[16]

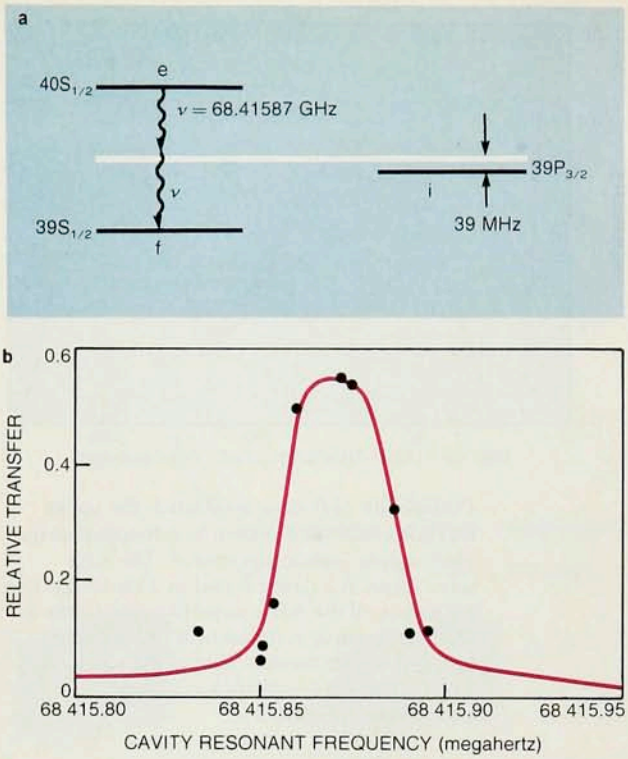
Other cavity QED effects

In addition to changing the radiation rate of atoms, atom–cavity coupling also induces energy shifts. This phenomenon can be understood as a frequency-pulling effect that occurs when an atomic "oscillator" interacts with a reactive cavity. Quantum mechanically, one can analyze it in terms of the effect of the cavity walls on the virtual-photon exchange processes. The metallic boundaries alter the modes of the vacuum field around the atom and affect not only real photon emission—that is, spontaneous emission rates—but also the virtual process responsible for radiative energy shifts.

A superficial analysis of the experiments on the inhibition of spontaneous emission could lead one to believe that suppressing the natural line width might give rise to arbitrarily narrow spectral lines and ultrahigh spectroscopic resolution. The fact that the atomic energy levels are at the same time shifted from their "free space" positions reveals that one is really carrying out spectroscopy on an atom "dressed" by the cavity vacuum, which is different from the atom "dressed" by the free-field vacuum. Precise experiments are now beginning to observe these effects. For example, experiments on individual trapped electrons are reaching the point where cavity-induced shifts are affecting measurements of the electron magnetic moment.[17]

The group at the MIT Spectroscopy Laboratory has observed frequency shifts caused by cavity effects at optical wavelengths, using barium atoms in a concentric optical resonator.[18] They saw changes in both the transition frequency and the linewidth caused by enhanced and inhibited spontaneous emission.

Cavity QED has potential applications to the study of nonlinear dynamics in small systems. A tenuous beam of atoms crossing a high-Q cavity one at a time is a highly nonlinear system with feedback—the field radiated by one atom reacts strongly back on the next atom. These are the ingredients required for chaos in a classical system. When solving the classical equation of motion for such a system, one finds chaotic behavior. Studying the quantum behavior of the system in the regime where the classical model exhibits bifurcations and chaos may provide insight into the connection between quantum mechanics and nonlinear classical mechanics.



Two-photon Rydberg maser.[15] The maser operates between two levels, *e* and *f*, of the same parity in rubidium. **a:** The cavity, tuned to exactly half the frequency of the *e*–*f* interval, enhances the two-photon process and inhibits the usually dominant one-photon emission toward the intermediate level *i*. **b:** Plot of the ratio of the lower-state population to the upper-state population as the cavity is tuned across the two-photon resonance. The peak indicates two-photon maser operation. **Figure 6**

The resonant-cavity-QED effects described above can be generalized to many-atom systems. Instead of coupling the atoms to the cavity one by one, it is possible to prepare samples of N identical Rydberg atoms in the cavity and study their collective behavior. Because of strong interatomic correlations, all the radiative phenomena occur much faster than they do with single atoms. Experimenters have observed[19] cooperative "superradiance" taking place within a time $(N\Gamma_0)^{-1}$ as well as collective Rabi oscillations involving a reversible exchange of energy between the field and the atomic sample at the enhanced rate $\Omega_{ef}\sqrt{N}$.

In the case of a strongly pumped two-level system, cavity effects can profoundly alter the dynamics of radiative relaxation and the population densities at equilibrium. A group at the University of Oregon has studied such effects theoretically and experimentally.[20]

Finally, cavity-QED experiments provide a link between microscopic and macroscopic physics. As the number of atoms and photons in the cavity is increased, the system evolves from one in which quantum fluctuations are dominant to one in which statistical fluctuations take over. This evolution involves such phenomena as quantum tunneling and quantum diffusion. The survival of purely quantum effects in a macroscopic system where fluctuation and dissipation are important is interesting, and simple atom–cavity systems are good candidates for studying it.

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