Observation of Cavity-Enhanced Single-Atom Spontaneous Emission

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It has been observed that the spontaneous-emission lifetime of Rydberg atoms is shortened by a large ratio when these atoms are crossing a high-Q superconducting cavity tuned to resonance with a millimeter-wave transition between adjacent Rydberg states.

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Spontaneous atomic emission inside an electromagnetic cavity is expected to occur at a rate different from the same process in free space.[1-4] If the cavity is resonant with a transition between two atomic levels, the partial spontaneous emission rate associated with the transition is multiplied by $\eta_{cav} = 3Q\lambda^3/4\pi^2 v$ where Q is the cavity quality factor, v its volume, and λ the transition wavelength. This effect, first discussed in the context of radio frequencies by Purcell in 1946,[1] is due to the change of the number of radiator modes per unit volume and unit frequency induced by the presence of the cavity. It can equivalently be understood as resulting from the interaction between the atom and its electric images reflected in the cavity mirrors. This effect has never so far been observed experimentally on a single-atom system.[5] In the optical domain, the usual open Fabry-Perot cavities have an effective volume v much larger than λ^3 so that, in spite of high Q's, η_{cav} is very small compared to unity. In the radio-frequency domain, on the other hand, it is relatively easy to realize low-order cavities with $v \simeq \lambda^3$ and high Q's corresponding to $\eta_{cav} \gg 1$. However, the free-space spontaneous emission rate Γ_0 is usually exceedingly small in this frequency range so that—even enhanced—a spontaneous emission process remains very difficult to detect.

In this Letter, we report the first observation of enhanced atomic spontaneous emission in a resonant cavity. The experiment has been performed with Rydberg atoms of Na excited in a niobium superconducting cavity resonant at 340 GHz. Taking advantage of the very strong electric dipole of these atoms (Γ_0 is unusually large on millimeter-wave transitions in Rydberg systems) and of the very good finesse of superconducting resonators, we have been able to observe a cavity-tuning-dependent shortening of the lifetime of atoms crossing, a few at a time, the resonant cavity.

Our experimental setup is represented in Fig. 1. Rydberg atoms in the $23S$ state are produced in a millimeter-wave Fabry-Perot resonator by stepwise excitation of a thermal beam of Na atoms with short (5 ns duration) dye-laser pulses (the collinear laser beams, perpendicular to the plane of Fig. 1, are not shown). The pulse repetition rate is 10 sec^{-1}. By changing of the laser intensities, the average number N of atoms excited by each pulse can be varied from a value of the order of one to several thousands. The cavity is made of two spherical niobium mirrors (diameter 20 mm, radius of curvature 26 mm) close to the confocal configuration at a distance $L \simeq 25$ mm from each other. By varying of L (with the help of the tuning screw shown in Fig. 1), the cavity is tuned to resonance with the transition towards the less excited $22P_{1/2}$ or $22P_{3/2}$ levels ($\nu_1 = 340.967$ or $\nu_2 = 340.396$ GHz and $\lambda_1 \simeq \lambda_2 \simeq 0.88$ mm). The mode sustained by the cavity has a Gaussian transverse profile with a waist $w = 1.9$ mm. The atoms are excited and remain close to an antinode position in the standing-wave pattern of the field while they drift in the cavity along a direction perpendicular to its axis. The average time they spend in the mode is $\Delta t = 2$ μs. The equivalent mode volume for this configuration is $v = \pi L w^2/4 = 70$ mm^3, corresponding to an enhancement factor $\eta_{cav} = 7.4\times10^{-4}Q$. Since the partial spontaneous decay rate from level $23S$ to the $22P$ state in free space is $\Gamma_0 = 150$ s^{-1}, the cavity enhanced rate is (in inverse seconds) $\Gamma_{cav} = \eta_{cav}\Gamma_0 = 0.11Q$, and the expected fraction of atoms transferred by enhanced spontaneous emission to the $22P$ state during Δt is $\Gamma_{cav}\Delta t = 2.2\times10^{-7}Q$. Quality factors of the order of 10^6 are thus required to perform the experiment. In order to obtain such high Q's, the mirrors are cooled below the niobium superconducting transition point (9.2 K) with the help of a liquid-helium cryostat shown in the upper part of Fig. 1. The operating temperature of the cavity, measured by a bolometer taped on the lower mirror, is $T = 5.7$ K. For this temperature the surface resistivity of the niobium at 340 GHz corresponds to a theoretical Q of several times 10^6.[6] In order to get

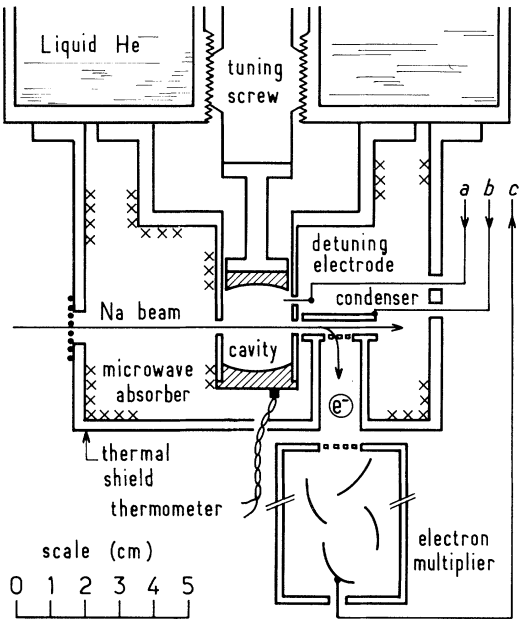

FIG. 1. Experimental arrangement.

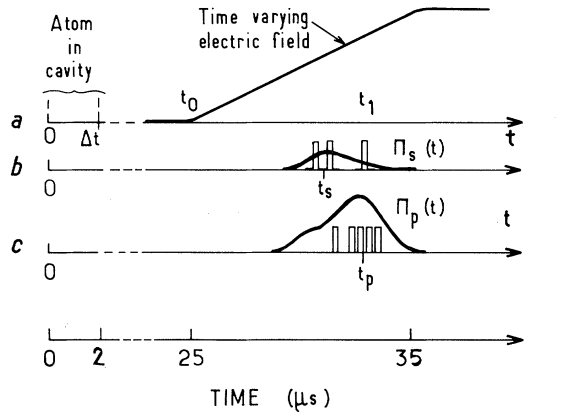

FIG. 2. Detection time sequence. *a*, Ionization field; *b*, detection of the 23*S* initial level; *c*, detection of the 22*P* final level.

as close as possible to this limit, extreme care has been taken to prepare the mirrors. They have been carefully polished, then annealed, and finally superficially cleaned by chemical reaction before being mounted in the vacuum chamber.[6] Before the superconducting transition is reached, the magnetic field in the cavity is cancelled, which avoids the trapping of spurious fields in the cavity mirrors.

Cooling the apparatus also has the important result of practically suppressing the effect of blackbody radiation on Rydberg atoms. The cavity and the atomic beam are surrounded by a cylindrical copper shield cooled to 7 K by contact with the Dewar. The laser and atomic beam windows are covered with a fine mesh providing a 10-dB attenuation for blackbody millimeter-wave radiation from outside. The inside of the shield is coated with black graphite paint absorbing the microwaves. The radiation inside the shield is thus thermalized at 7 K. Moreover, the background radiation in the shield cannot couple into the cavity, whose Q is not diffraction limited. The radiation in the cavity mode is thus thermalized at the temperature of the mirrors ($T = 5.7$ K), which corresponds to an average photon occupation number $\bar{n} = [\exp(h\nu/k_B T) - 1]^{-1} = 0.06$. Stimulated emission by blackbody radiation in the cavity mode is thus negligible compared to spontaneous emission ($\bar{n} \ll 1$).

After they have left the cavity, the atoms are state analyzed by ionization with an electric field applied between two 4-mm-distant parallel condenser plates (Fig. 1, line *b*). The resulting electrons are detected by an electron multiplier (Fig. 1, line *c*). The detection procedure is time analyzed in Fig. 2. The atoms pass in front of the entrance window of the electron multiplier around time $t_1 = 33$ μs. (The time origin corresponds to the laser excitation.) A voltage increasing linearly with time is applied on the plates, starting at time $t_0 = 25$ μs. It reaches its maximum 1-kV value within about 10 μs (see Fig. 2 curve *a*). The ionization mechanism of the Rydberg atoms in the time-varying electric field is a complicated process depending upon the Stark structure of the levels. For our purpose, it is only important to realize that the ionization occurs on the average at a lower field for the 23*S* state than for the more tightly bound 22*P* level. As a result, the probabilities $\Pi_S(t)$ and $\Pi_P(t)$ of detecting an electron released by an atom in level 23*S* or 22*P* are spread over intervals centered around times t_S and t_P shifted with respect to each other by about 1 μs (Fig. 2, curves *b* and *c*). A transition occurring between the 23*S* and 22*P* states in the cavity results in a change of the electron detection probability for the corresponding atom from the $\Pi_S(t)$ to the $\Pi_P(t)$ distribution. In order to relate the number of electrons to the number of atoms having actually crossed the cavity, one has to take into account a transmission factor $k = 0.6$ due to velocity spreading of the atoms (not all atoms are in front of the detector window at time t_1) and also spontaneous emission processes during the time t_0 towards all lower-lying levels with principal quantum numbers smaller

than 22.[7] The atoms decaying into these more tightly bound states are not ionized in the 1-kV electric field ramp and are not detected at all. The lifetimes of the $23S$ and $22P$ states being[8] $\tau_S = 14.5\ \mu s$ and $\tau_P = 110\ \mu s$, the fractions of atoms reaching the detector are respectively $\int \Pi_S(t)\,dt = k \exp(-t_0/\tau_S) \simeq 0.1$ and $\int \Pi_P(t)\,dt = k \times \exp(-t_0/\tau_P) \simeq 0.4$. Note that the $22P$ state's much longer lifetime is an asset for our detection procedure, any atom having spontaneously radiated on the $23S \rightarrow 22P$ transition inside the cavity being detected with a probability four times as large as an atom remaining in the $23S$ state. The electron pulses detected by the electron multiplier are sent to a Tektronix R7912 transient analyzer interfaced to a LSI 11 computer. This computer allows us to measure the average number of atoms excited in the cavity by each laser pulse, and to store and process the time-resolved ionization signals.

The experiment is performed in the following way: (i) Using strong laser beams, we excite first a large number of atoms per pulse ($N \gtrsim 1000$). The cavity being out of resonance, the resulting electron signal has a time dependence characteristic of the $\Pi_S(t)$ distribution, which we determine in this way. (ii) We then proceed to tune the cavity into resonance with the $23S \rightarrow 22P_{1/2}$ or $23S \rightarrow 22P_{3/2}$ transition by translating the upper mirror. The atom-to-cavity frequency matching is monitored by observing at resonance a strong radiative transfer from the $23S$ to the $22P$ level, corresponding to a cavity-assisted superradiance effect involving the large-N atom sample.[9] At resonance, all atoms leave the cavity in the final $22P$ state which results in a fourfold increase of the atomic signal as compared to (i). The electron signal time dependence is now proportional to the $\Pi_P(t)$ distribution, which we determine in this way. The width of the cavity resonance corresponds to a very small mirror translation $\delta L = L/Q \simeq 200$ Å, that is to a rotation of only 3×10^{-4} rad of the tuning screw. The tuning procedure is thus very critical and the resonance condition impossible to maintain over more than a few minutes because of cavity length instabilities. (iii) In order to study single-atom effects and to avoid collective atomic behavior in the cavity, we then reduce the laser intensities to a level such that the average number of detected electrons is much less than one per laser pulse. At such low counting rates, the number of atoms crossing the cavity at a time is of the order of unity and the radiative transfer is of course not observable on single laser shots. We average 200 time-resolved ionization signals with the cavity on resonance and compare to the corresponding average obtained with the cavity off resonance. On- and off-resonant signals are alternated from one laser shot to the next. Since it is impossible to realize this fast on and off tuning mechanically, the cavity length is not modified, but we slightly Stark-shift the atomic levels by applying immediately after every other laser pulse a 70-V voltage to a small electrode close to the cavity mode (Fig. 1, line a).

The experimental results are displayed in Fig. 3. In Fig. 3, traces a, b, and c have been obtained with an average number of detected electrons in the $23S$ state (nonresonant case) equal to 0.35, 0.2, and 0.13, respectively, corresponding to an average of 3.5, 2, and 1.3 atoms excited per pulse (with a Poisson distribution around this average). Figure 3, trace c, for example, corresponds to events in which 0, 1, 2, or 3 atoms at most are crossing the cavity at a time. The full-line resonant curves have a mixed $\Pi_{SP}(t)$ shape which we analyze as a superposition $\Pi_{SP}(t) = (1-x)\Pi_S(t) + x\Pi_P(t)$, x being the measured fraction of atoms transferred by the cavity-enhanced spontaneous emission process. We get $x = 0.15 \pm 0.05$ which yields $\Gamma_{cav}\,\Delta t = 2.2 \times 10^{-7} Q = -\ln(1-x) = 0.16$ and $Q \simeq 7.5 \times 10^5$. This Q value

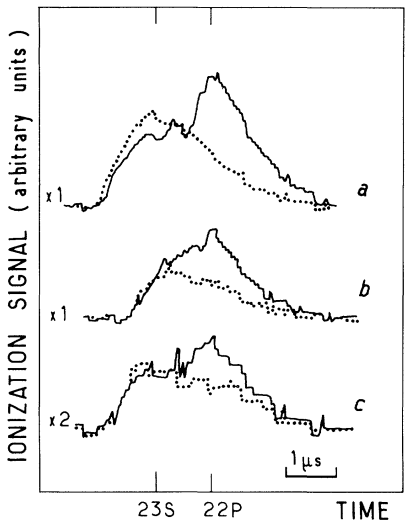

FIG. 3. Cavity-enhanced spontaneous emission signals. Dotted line, off-resonant cavity; full line, resonant cavity. The average numbers of atoms in the cavity are respectively 3.5, 2, and 1.3 in traces a, b, and c. Traces a and c correspond to $23S \rightarrow 22P_{3/2}$, trace b to $23S \rightarrow 22P_{1/2}$.

should be considered as a lower limit since it is difficult to ensure perfect cavity stability during the data acquisition. (Also one cannot exclude the effect of submegahertz Stark shifts, introduced by small electric fields in the cavity, which would detune the atoms in a time smaller than Δt and make the Q appear smaller than it actually is.)

It is important to realize that the condition $Nx < 1$ ($N = 1, 2, 3, \ldots$) implies that the probability of having more than one photon per pulse emitted in the cavity mode during Δt is very small. As a result, the radiative coupling between atoms is negligible even if two or three atoms are prepared in the cavity by a single laser pulse. We are thus testing in this experiment *single-atom* radiative properties. With longer interaction times, however, the collective coupling of the atoms would take place and the evolution rate would become N dependent, exhibiting cavity-assisted superradiant effects involving two or three atoms. In this respect, the effect described in this Letter can be considered as the limiting case of a transient maser approaching threshold with only one or two atoms in the inverted medium.

We have shown by this experiment that the partial spontaneous emission probability on the $23S \rightarrow 22P$ transition in Na is increased in a high-Q cavity from its free-space value $\Gamma_0 = 150\ s^{-1}$ up to $\Gamma_{cav} = 8 \times 10^4\ s^{-1}$. Let us notice that this enhanced rate is still 35 times smaller than the damping rate $2\pi\nu/Q = 2.8 \times 10^6\ s^{-1}$ of the field in the cavity. In other words, the photon emitted in the mode is absorbed in the mirrors much faster than the atom decays. This is the justification for describing the emission as an irreversible process. With a tenfold increase in Q, Γ_{cav} and $2\pi\nu/Q$ would become of the same size and the emitted photon would be stored in the cavity long enough for the atom to be able to reabsorb it. This would correspond to a regime of quantum mechanical oscillations between a two-level atom and a single electromagnetic field mode[4] which should be observable with an improved version of our setup.

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

[2]G. Barton, Proc. Roy. Soc. London, Ser. A 320, 251 (1970); P. Stehle, Phys. Rev. A 2, 102 (1970); P. W. Milonni and P. L. Knight, Opt. Commun. 9, 119 (1973); M. R. Philpott, Chem. Phys. Lett. 19, 435 (1973).

[3]D. Kleppner, Phys. Rev. Lett. 47, 233 (1981); S. Haroche, C. Fabre, J. M. Raimond, P. Goy, M. Gross, and L. Moi, J. Phys. (Paris), Colloq. 43, C2-265 (1982).

[4]S. Haroche, P. Goy, J. M. Raimond, C. Fabre, and M. Gross, Philos. Trans. Roy. Soc. London, Ser. A 307, 659 (1982).

[5]The influence of a single conducting surface on the fluorescence of a molecule has been observed; see K. H. Drexhage, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1974), Vol. 12, p. 165. This effect, which involves the molecule's interaction with a single image, is nonresonant and very small compared to the one described here.

[6]V. Nguyen Tuong, L. Wartski, P. Goy, J. M. Raimond, M. Gross, and S. Haroche, to be published.

[7]After t_0, the electric field ramp mixes the short-lived $23S$ state to the much longer-lived $22P$ one, so that spontaneous emission loss during time $t_1 - t_0$ is negligible.

[8]We consider of course here the global lifetimes, not to be confused with the partial lifetime Γ_0^{-1} on the $23S \rightarrow 22P$ transition, which is the only one altered by the cavity ($\tau_S^{-1} \gg \Gamma_0$). For a determination of these lifetimes, see J. F. Gounand, J. Phys. (Paris) 40, 457 (1979); W. P. Spencer, A. G. Vaidyanathan, D. Kleppner, and T. W. Ducas, Phys. Rev. A 24, 2513 (1981).

[9]M. Gross, P. Goy, C. Fabre, S. Haroche, and J. M. Raimond, Phys. Rev. Lett. 43, 343 (1979).