Rydberg Atoms And Radiation In A Resonant Cavity II. Experiments

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RYDBERG ATOMS AND RADIATION IN A RESONANT CAVITY II. EXPERIMENTS

C. Fabre, S. Haroche, J.M. Raimond, P. Goy, M. Gross and L. Moi

*Laboratoire de Physique de l'Ecole Normale Supérieure, 24 rue Lhomond,
75231 Paris Cedex 05, France*

Résumé.- Nous présentons différentes expériences mettant en jeu des atomes de
Rydberg en interaction avec une micro-onde résonnante à l'intérieur d'une ca-
vité, permettant de vérifier différents aspects de la théorie présentée dans
l'exposé précédent. Nous décrivons d'abord des expériences d'émission super-
radiante des états de Rydberg dans la cavité   (effet maser transitoire
à atomes de Rydberg).L'évolution du système est détectée soit sur les popula-
tions par la technique d'ionisation sélective par champ, soit sur le champ micro-
onde par détection hétérodyne. Des seuils très bas de l'émission maser, jusqu'
à 100 atomes, ont été observés. Nous décrivons ensuite quelques aspects de
l'absorption collective de rayonnement par les atomes très excités, en parti-
culier l'observation de la nutation de Rabi sous l'effet d'un faible champ
extérieur, l'étude du rendement quantique de notre système considéré comme un
compteur de photons millimétriques et enfin la mise en évidence d'aspects inat-
tendus dans l'absorption de rayonnement du corps noir dans la cavité.

Abstract.- We present various experiments involving Rydberg atoms interacting
with resonant microwave electromagnetic fields inside a cavity, allowing to
check different features of the theory presented in the previous paper. We
first describe experiments about superradiant emission by Rydberg states in
the cavity, also called transient Rydberg atoms maser; the evolution of the
system is detected either on the populations by selective field ionization
technique or on the microwave field itself by heterodyne detection. Very low
thresholds, down to 100 atoms, for the maser emission have been observed.
We then describe various aspects of collective absorption of radiation by very
excited atoms including the observation of Rabi nutation in a small applied
resonant field, the study of the quantum efficiency of this set-up considered
as a microwave photon counter, and finally the unexpected characteristics of
the blackbody radiation absorption in the cavity.

In this second paper, we present various experiments on Rydberg states inter-
acting with resonant millimetric electromagnetic waves inside a cavity. These expe-
riments show various interesting physical effects calculated in the previous paper
and provide a test for the theoretical models. In a first paragraph, we will des-
cribe the experimental techniques which are common to all the experiments that will
be presented afterwards. We will devote the second paragraph to an experimental
study of the different characteristics of the collective Rydberg atom transient
emission in the cavity (the "Rydberg atom maser"). In the last paragraph, we will
describe several experiments involving the absorption of radiation by very excited
states in the cavity and showing the collective behaviour of the Rydberg atoms in
this absorption process.

1. Experimental techniques
    1.1. Preparation of the physical system
    In order to prepare the physical system which has been theoretically studied
in the previous paper, i.e. a set of 2-level Rydberg atoms in resonant interaction
with an electromagnetic mode inside a cavity, we use the experimental set-up which

is schematized on Figure 1 :

- Two simultaneous laser pulses populate,by a stepwise excitation, a given Rydberg state |nS > (n $\approx$ 30) of alkali atom (sodium or cesium) in a thermal beam. The number N of very excited states created by each laser pulse can be varied from 1 to 10^6. Due to the long lifetimes of such states (several tens of μs), they drift at thermal velocities along the atomic beam over several centimeters before decaying to lower lying states.
- The Rydberg atoms are created in an antinode of the standing wave of the Gaussian mode
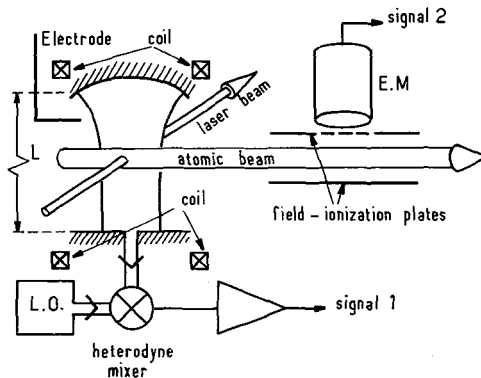

Figure 1 : Scheme of the experimental set-up

existing in a semi-confocal Fabry-Perot cavity, the axis of which is perpendicular to the atomic beam. The resonant frequency of the cavity can be adjusted by varying the distance L between the two mirrors. The cavity typical characteristics are the following : length L : $\sim$ 1 cm; waist w_0 of the Gaussian mode : $\sim$ 3 mm; volume $\mathcal{V}$ of the mode in the cavity : $\sim$0.1 cm^3; quality factor Q around 150 GHz : 10^4; damping time of the mode in the cavity $Q/\omega = T_{cav}$ $\sim$ 9 ns (which is much less than the transit time T $\sim$ 3 μs of the atoms inside the Gaussian mode). The cavity can be fed through a wave guide by an external mm source (carcinotron or frequency multiplied klystron).

- For most experiments, the cavity is tuned to the Bohr frequency of the transition $nS\frac{1}{2} \rightarrow n'P\frac{1}{2}$ (n' = n or n - 1), so that two transitions $|nS\frac{1}{2} \; m_J = +1/2 > \rightarrow$ $|n'P\frac{1}{2} \; m_J = -1/2 >$ and $|nS\frac{1}{2} \; m_J = -1/2 > \rightarrow |n'P\frac{1}{2} \; m_J = +1/2 >$ interact independently with the σ_+ and σ_- polarizations of the electromagnetic field mode inside the cavity (the "quantization" Oz axis being chosen along the cavity axis). For some experiments, it has been necessary to study a unique two-level system interacting with the cavity mode. For that purpose, two coils coaxial to the cavity can be used to apply a static magnetic field B_0 along the z-axis and split apart the two Zeeman components of the transition. If the field is large enough ($B_0 \gtrsim$ 50 G), the Zeeman splitting $\sim$ 100 MHz is larger than the width of the cavity mode, so that the cavity can be tuned to resonance with a single two-level-atom system.

1.2. Detection of the evolution

Two techniques have been used to detect the evolution of the system atom + field, the first one on the field, and the second on the atoms :
(i) To monitor the electromagnetic field at the output of the cavity, we use an heterodyne detector ([1]) (signal 1 in Fig. 1) : it consists in a low noise Schottky diode which mixes the signal to measure and the output of a local oscillator (stabilized carcinotron). The low frequency beatnote ($\sim$ 100 MHz) is then amplified, filtered, monitored, and eventually rectified and averaged over several laser shots. This technique allows us to record the whole evolution of the system for each laser pulse without perturbation ,   but is restricted

to rather "intense" signals (number N of excited atoms $\gtrsim 10^5$ which corresponds to a signal of 10 pW during a few hundreds nanoseconds). Furthermore, the narrow bandpass filter used to reduce the thermal noise on the beat signal limitates the time response of the detection to roughly 80 ns.

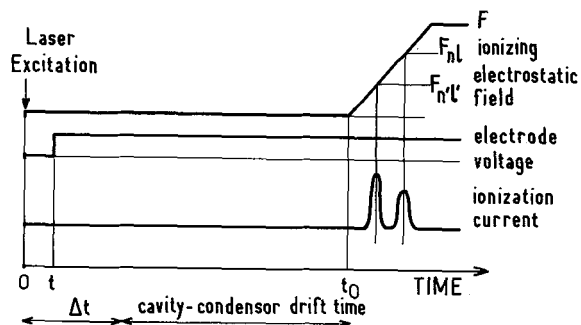(ii) To detect the evolution of the Rydberg level populations, we make use of the

Figure 2 : Time sequence of different experimental parameters. Δt is the time of flight of the atoms in the cavity mode. The atom-cavity interaction is stopped at time t by the voltage applied on the electrode. The ramp of electric field applied from time t_0 ionizes at different times the different Rydberg levels.The resulting ionization current displays peaks corresponding to the different Rydberg states populated at time t_0.

now well-known selective electrostatic field ionization technique (signal 2 in Fig. 1) : at the exit of the Fabry-Perot cavity, the atoms enter a plane condenser inside which an electrostatic field F is applied. Electrons created by ionization at the center of the condenser can be detected through a mesh by an electron-multiplier. Figure 2 shows the sequence of events for each laser pulse : at time t = 0, the Rydberg atoms are created inside the cavity and interact with the electromagnetic field mode during a time Δt. They drift afterwards in the condenser and are submitted from time t_0 to a ramp of electric field F(t). The thresholds F_{nS} and $F_{n'P}$ are reached at slightly different times. The ionisation current exhibits two peaks as a function of time, the areas of which yield the populations of the levels nS and n'P at time t_0 (when the gain of the e.m. is known). Since the effective lifetimes and the thermal transfer rates of the two levels are   known, we can deduce the populations just after the end of the interaction between the atoms and the cavity. To know the populations of the two levels at any time t < Δt during the evolution inside the cavity, we use the following method : at time t, a fast rising voltage is applied between an electrode E (see Fig. 1) and the grounded cavity : this voltage induces an inhomogeneous Stark shift such that the transition frequency between levels nS and n'P is no longer resonant with the cavity : the interaction atoms-cavity stops at this time t and the system is "frozen" in its evolution. By varying t we can sample the evolution of the atomic system.

The whole detection system is interfaced with a computer LSI11 which computes on line the populations of the levels, and in order to get rid of the pulse-to-pulse fluctuations of the laser excitation,records only the ionization signal corresponding to a given number N (within a few %) of initially excited Rydberg levels. This sampling technique is very convenient and can be used even for very low N numbers of excited atoms, since the field ionization detection can count one by one the Rydberg atoms arriving inside the condensor.

2. Experimental study of Rydberg state superradiance in a cavity (Rydberg masers)

2.1. General characteristics [2][3][4][5]

A first class of experiments consists in exciting N Rydberg atoms in an $|nS>$ state inside the cavity and in tuning the cavity to resonance with a transition linking this state to a lower lying $|n'P>$ state. Usually n' = n-1, i.e. n'P is the closest P level of larger binding energy, but it can be also n-2, n-3 in some cases. In the Bloch vector representation, it corresponds to an initial Bloch angle $\theta = 0$. If N is large enough, the superradiant decay to the $\theta = \pi$ final state can occur before the atoms leave the cavity. The energy of the cavity electromagnetic mode is then temporarily increased by the coupling with the excited atoms : such an effect can be named transient Rydberg state maser. According to the actual characteristics of the cavities we have used, we have been always, so far, in the situation where $\omega/Q > T_R^{-1}$, which corresponds, according to the discussion of paragraph 6 in the previous paper, to an overdamped, non oscillatory, transfer of population to the final P state. The microwave energy I(t) is emitted by the system in a brief burst of radiation, following the well known hyperbolic secant form :

$$I(t) = \left[2 \ T_R \ \cos h \ \left(\frac{t - t_D}{2T_R} \right) \right]^{-2}$$

with t_D, delay time of the maser pulse,   given by equation (10) of the previous paper.

Two different detection techniques we have described in the previous paragraph can be used to obtain different informations on this physical system :

(i) The detection of the final populations of the two levels involved in the transition by field ionization technique has provided the first experimental evidence for the Rydberg state maser and yields valuable informations on the thresholds N_S of the maser action. Figure 3 displays the ionization current delivered by
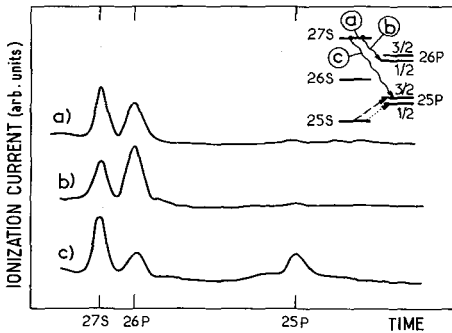
Figure 3 : Ionization signal showing maser action on different transitions when 27S state has been initially populated : a) untuned cavity; b) cavity tuned on 27S - 26S$\frac{1}{2}$ transition; c) cavity tuned on 27S - 25P$\frac{3}{2}$ transition

the electron multiplier as a function of time for different tuning positions of the cavity. The 27S level has been initially populated by the laser. Trace a) corresponds to an untuned cavity : we see the peaks corresponding to levels 27S (populated by the laser) and 26P (populated by thermal background outside the cavity during the transit time between excitation and detection. Trace b) corresponds to a cavity tuned on the 27S - 26P$\frac{1}{2}$ transition frequency (λ = 1.47 mm) and exhibits a strong enhancement of the 26P population, characteristic of the maser emission on this transition. When the cavity is now tuned to the 27S - 25P$\frac{3}{2}$ transition frequency

(λ = 0.466 mm), we observe a new peak on the ionization signal, corresponding to the 25P level. By this technique, we have put in evidence maser action on a large number of transitions between Rydberg states, ranging from 135 GHz to 1.44 THz.
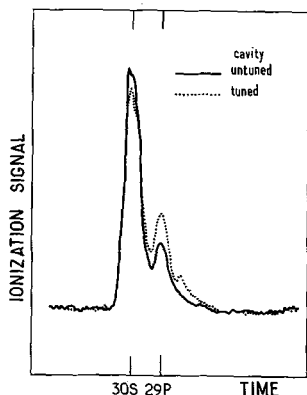

Figure 4 : Ionization signal showing maser action on 30S - 29P$\frac{3}{2}$ transition in a high finesse cavity with only 100 (± 50) excited atoms.

By determining the minimum number of excited atoms which yield an observable cavity-dependent population transfer, we can measure the threshold for such a maser emission. This threshold can be shown to be inversely proportional to the finesse $f = Q \left(\frac{\lambda}{2L} \right)$ of the cavity. Figure 4 shows a Rydberg atom maser near the threshold in a highly reflective cavity (f = 1600) : a difference in the population transfers between levels 30S and 29P$\frac{3}{2}$ ($\nu \sim$ 145 GHz) is clearly observable for a number of excited atoms as low as 100 (± 50). These very low thresholds are charac-teristics for maser action between Rydberg states and are to be compared to the thresholds of the order of 10^9 atoms for usual masers in the same frequency domain. By improving further the cavity mirrors reflectivity, we hope we will be able to obtain a collective emission of radiation by a very small number of Rydberg atoms, of the order of 1, for which the "single atom effects", outlined in paragraph 3 of the previous paper will be observable.

(ii) The detection by heterodyne techniques of the microwave signal emit-ted by the atoms has the advantage of giving the pulse shape and the delay of the emission. The left part of Figure 5 gives an example of such measurements, corres-
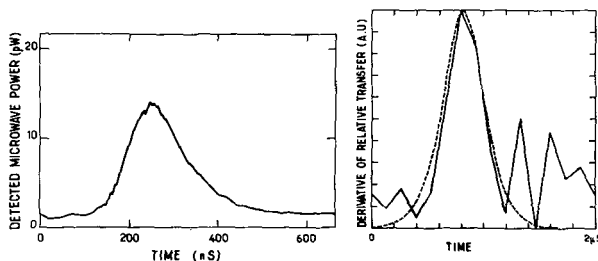

Figure 5 : Time evolution of the maser pulse : a) detected on the microwave by heterodyne mixing technique; b) detected by the "frozen evolu-tion" field ionization technique.

ponding to a peak intensity of 10 pW, delivered during a time of 100 ns. The time labelled 0 corresponds to the laser excitation time. The pulse shape and the average delays measured by this way can be shown to be in good agreement with the theoretical predictions ([4]).

        (iii) The right part of Figure 5 gives a typical signal recorded by the computer with the "frozen evolution" field ionization detection technique. The y-axis of this figure is in fact the time derivative of the population transfer, which is proportional to the field emitted by the atoms. This signal recorded on laser shots gives actually an average evolution of the maser corresponding to a given N value of excited atoms. It exhibits a bell-shaped behaviour typical of superradiance, and has been recorded with N = 50.000 only on the $30S \rightarrow 28P\frac{1}{2}$ transition (ν = 457.5 GHz).

2.2. Experimental study of the maser emission statistics

        A specific feature of the Rydberg state maser is the large pulse to pulse variations of the maser pulse characteristics, even when the number of initially excited atoms is kept constant. As explained in the previous paper, these fluctuations are nothing else but the amplification of the photon fluctuations during the early stages of the maser evolution from its unstable initial state. The "frozen evolution" field ionization detection technique, described in § 1.2, has enabled us to study experimentally the statistics of these fluctuations during the whole evolution of the system, and to quantitatively check with a good accuracy the formula (11) of the previous paper, giving the probability P(n,t) of emitting n photons between times 0 and t. The corresponding results will be presented in details elsewhere.

3. Experimental study of resonant absorption of radiation by Rydberg atoms

        Another series of physical effects is expected when one excites N Rydberg atoms in an |nS > state inside the cavity and simultaneously tune the cavity to resonance with the upward transition nS → nP. In this situation, the Bloch vector is initially put in the $\theta = \pi$ position, which is also the only stable permanent position of the vector. No spontaneous irreversible transfer to the other state of the transition can occur, and the only observable effects are then consisting in collective absorption of radiation by the atoms in interaction with the cavity mode.

3.1. Rabi nutation in Rydberg levels ([6])

        Let us first fill the cavity mode by an external resonant field of strength $\&_1$. The Bloch vector undergoes a Rabi nutation, resulting in a periodic variation of the populations of levels nS and nP, which can be recorded by the "frozen evolution" field ionization technique.

Figure 6 : Relative transfer from level 30P to level 30S as a function of time when a small external microwave field is applied : a) Rabi nutation when the number of excited atoms N is 300; b) radiative damping of this nutation when N $\approx$ 2000
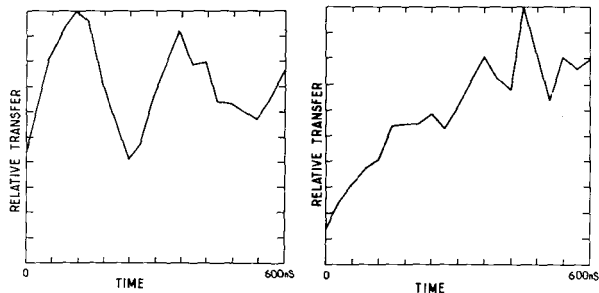
The left part of figure 6 gives as an example the ratio of the population of the 30P to 30S states of sodium as a function of time t, when N $\approx$ 300 atoms have been excited in the lower 30S state at time t = 0. The modulation of the transfer at the Rabi frequency $\nu_1 = \dfrac{d\mathcal{E}_1}{\hbar}$ is clearly observable. In this example, $\mathcal{E}_1 =$ 3.7 10^{-3} V/cm and ν_1 = 2.2 MHz. Due to the large dipole matrix element d, such a low field, corresponding to only 2000 millimeter photons stored in the cavity, is enough to induce a high frequency Rabi nutation. This kind of experiment is very convenient to calibrate in an absolute way the field in the cavity when one knows the electric dipole matrix elements.

But, when the Bloch-vector is put in the θ = 0 position at half the Rabi period, the collective superradiant spontaneous damping towards the $\theta = \pi$ position competes with the Rabi nutation. As a result, when the number N of excited atoms is high enough so that T_R is less than the Rabi period $1/\nu_1$, the Rabi oscillation is destroyed by the collective damping. Experimentally, for N > 2000, the Rabi nutation can be no longer observed (right part of Fig. 6).

This kind of collective damping broadening effects must be avoided when one performs very high resolution spectroscopy in cavities, which restricts the number of excited atoms to be used in a spectroscopy experiment.

With a cavity having a higher finesse, and hence a longer T_{cav} , it will be

possible to record similar evolutions corresponding to a very low number of excited atoms interacting with a very small number of photons present in the mode. We hope to be able to eventually observe "single atoms effects" such as those described in § 3 of the previous paper.

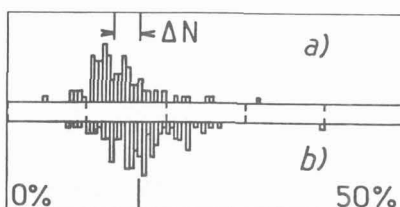### 3.2. Absorption of very small amount of radiation

Figure 7 : Histogram of relative transfers from level 30S to level 30P$\frac{1}{2}$: a) without external field; b) with an applied field of 3 10^{-14} w; an increase ΔN of the mean value of transfers is clearly appearing.

The previous experiment can be resumed with a strongly attenuated microwave field. Figure 7-b gives the histogram of transfers ΔN from level 30S to level 30P$\frac{1}{2}$ for N $\approx$ 1200 atoms and for a microwave power of 0.04 pW (4×10^8 photons/s at 134 GHz, i.e. 1000 photons impinging on the atoms at each pulse). Figure 7-a gives the same histogram without external microwave. The mean value   of the transfers clearly shifts from value 17% to the value 13%, this last value corresponding to the thermal background at room temperature. The difference ΔN = 4% corresponds to 50 photons   absorbed   by   the   atoms   , which yields a quantum efficiency η of the detection equal to 50/1000 = 5%. Such a quantum efficiency is quite good for this spectral domain and shows the advantage of using Rydberg atoms as mm wave detectors

3.3. Collective absorption of blackbody radiation ([7])

Another set of experiments has been performed, dealing with the absorption of thermal radiation background by Rydberg atoms. More precisely, we have observed the modification of this absorption when the cavity is tuned to resonance with the $30S\frac{1}{2}$ - $30P\frac{1}{2}$ transition (ν = 134 GHz), the $30S\frac{1}{2}$ state being initially populated by the laser. We measured here, by field ionization technique, the populations of S and P levels at the end of the coupled evolution atom + cavity. When the cavity is off-resonant, one observes, at room temperature, a thermal transfer of roughly 10% from $30S\frac{1}{2}$ level to $30P\frac{1}{2}$, corresponding to the absorption of blackbody radiation by the atoms in the modes transverse to the cavity axis and outside the cavity during the drift from the cavity to the detection region (fig. 8-a). When the cavity is tuned to resonance, an increase ΔN of the number of transferred atoms is observed.
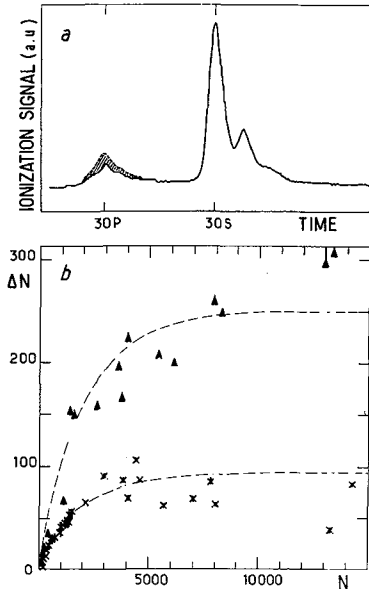

Figure 8 : a) ionization signal showing the variation of population transfer from 30S to 30P level when the cavity is untuned (full line) and tuned to resonance (dashed line). b) variation of transfer ΔN as a function of the number N of excited atoms; crosses : experiment performed at 300°K; triangles : same experiment with a heated field mode (the experimental uncertainty on N is around 25%)

Figure 8-b gives a plot of ΔN, averaged over 300 events, as a function of the total number of excited atoms N. We see that, in agreement with the theory of the previous paper, ΔN saturates to a value of the order of $2\bar{n} = 2k_B T / \hbar\omega$ for large N values. The lower part of Figure 8-b gives the limit 80 ± 20 absorbed photons for the set of 2 two-level systems coexisting in the $30S\frac{1}{2} \rightarrow 30P\frac{1}{2}$ transition. When a magnetic field is applied in the cavity, in the way described in paragraph 1.2, so that a single two-level atom system interacts with the cavity mode, we have checked that the asymptotic value is divided by roughly a factor 2; the formula $\Delta N = g\,\frac{kT}{h\nu}$ where g = 2 or 1 is the degeneracy factor, gives a value in agreement with the measured one, within the error bars. This is an experimental evidence that the system of N excited atoms behaves as a single collective system since it can absorb only $\frac{kT}{h\nu}$ thermal photons per two level system and not N/2 , as would be expected if the atoms were coupled independently from each other to the cavity.

This system provides an absolute thermometer to measure the effective temperature of the cavity mode. The upper part of Fig. 8-b has been obtained when this mode was heated by a hot tungsten wire (T $\simeq$ 1600°K) coupled to the cavity mode. One sees clearly an increase of the maximum number of transferred atoms ΔN=270±60

(no magnetic field was present in this case), the previous formula with g = 2 yields a value $T_{rad} \approx 900°K$, which means that the radiation reaches a temperature equal to about half the hot wire one, a result consistent with estimates based on simple cavity coupling models.

4. Conclusion

This brief survey of experimental results has allowed us to emphasize some striking features of Rydberg states in interaction with an electromagnetic wave in a cavity; in particular it has been shown that the collective radiative decay of the whole sample plays a very important part in different experiments : Rydberg maser of course, but also Rabi nutation in an external field and absorption of blackbody radiation.

References

[1] GOY P., Int. J. of Infrared and Millimeter waves, 3 (1982), 221

[2] GROSS M., GOY P., FABRE C., HAROCHE S., RAIMOND J.M., Phys. Rev. Letters, 43 (1979), 343

[3] MOI L., FABRE C., GOY P., GROSS M., HAROCHE S., ENCRENAZ P., BEAUDIN G., LAZAREFF B., Optics Commun. 33 (1980), 47

[4] MOI L., GOY P., GROSS M., RAIMOND J.M., FABRE C., HAROCHE S., submitted to Phys. Rev. A (1982)

[5] GOY P., MOI L., GROSS M., RAIMOND J.M., FABRE C., HAROCHE S., submitted to Phys. Rev. A (1982)

[6] HAROCHE S., GOY P., RAIMOND J.M., FABRE C., GROSS M., to be published Proc. Roy. Soc. (1982)

[7] RAIMOND J.M., GOY P., GROSS M., FABRE C., HAROCHE S., Phys. Rev. Letters, 49 (1982), 117.