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Vacuum Rabi Splitting Observed on a Microscopic Atomic Sample in a Microwave Cavity.

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Abstract. – We have observed the vacuum Rabi splitting on a beam of Rydberg atoms resonantly coupled to a high-Q superconducting cavity mode. The splitting is resolved down to five atoms on average inside the cavity and only three atoms resonant with the mode.

The frequency degeneracy of two oscillators resonant with each other is removed by their coupling. The symmetric and antisymmetric combination of oscillation amplitudes constitute normal modes with eigenfrequencies split apart by an amount proportional to the geometric average of the oscillators coupling parameters. The frequency splitting also represents the rate at which the two oscillators exchange their energy. This effect has innumerable consequences in classical and quantum mechanics and electrodynamics. It has recently received renewed attention in the context of cavity QED [1]. In this case, one oscillator is a small collection of N identical atoms, the other is a resonant mode of a high-Q cavity. The coupled atom-cavity system is weakly excited by a tuneable field probe and its excitation is recorded as a function of the probe frequency. This excitation is not resonant at the bare atom or cavity frequency, but at the split frequencies of the «dressed» atom-field system. For N two-level atoms, motionless and equally coupled to the mode, the splitting is predicted [2] to be $2\Omega_0\sqrt{N}$, $2\Omega_0$ being the «single atom vacuum Rabi frequency». A challenging experimental goal is to record the spectrum down to $N=1$ and to observe the splitting predicted in [3], which corresponds to the coherent coupling of a single atom with a single photon. Dual interpretations of the N-atom vacuum Rabi splitting effect can be given: i) the atomic sample is a refractive medium with a complex index which has the effect to separate the cavity mode into two components [4] and ii) the cavity vacuum fluctuations induce a resonant dynamical Stark effect on the atoms, with the result of splitting their resonance line by an amount proportional to the collective atomic-dipole amplitude [1].

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Vacuum Rabi splitting has been observed in the optical domain on the transmission of Fabry-Perot resonators containing a small number of resonant sodium [5] or barium [4] atoms. In this letter, we report its first observation on microscopic samples of Rydberg atoms coupled to a microwave superconducting cavity. We have observed the normal mode splitting with as few as 5 atoms at a time coupled to the cavity, 3 of which only being resonant with the cavity mode. This experiment is a new illustration of the extreme sensitivity of Rydberg atoms to vacuum field effects. A notable difference with the optical normal mode splitting experiments resides in the much longer life times of the atomic and field excitations (ten and hundred microsecond ranges, respectively, instead of tens of nanoseconds). The observation of the vacuum Rabi splitting effect on Rydberg atom systems opens the way to interesting experiments performed on photons «trapped» over a long period of time in a cavity and probed by a beam of radiatively long-lived atoms [6].

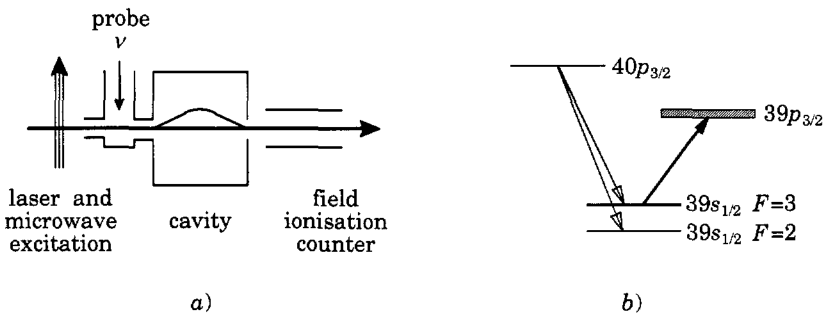

Fig. 1. – *a*) Sketch of experimental set-up: the atomic beam (horizontal arrow) crosses the excitation zone, the cavity and the field ionization detection region. The probing field is injected in the cavity through the same wave guide as the atoms. The sine arch variation of the electric-field amplitude is indicated. *b*) Diagram of relevant energy levels. The microwave preparation step populating the $39S_{1/2}$, $F = 2$ and 3 hyperfine levels is indicated by the downward arrows. The $39P_{3/2}$ hyperfine structure is unresolved. The cavity mode resonantly couples the $39S_{1/2}$, $F = 3$ and $39P_{3/2}$ levels (upward arrow).

Our experimental set-up is sketched in fig. 1*a*). A beam of Rb atoms crosses a cylindrical superconducting Nb cavity, cooled to 1.7 K. The cavity (length: 7.5 mm, diameter: 7.75 mm) sustains the TE_{121} mode whose amplitude varies along the cavity axis as a sine arch. The r.m.s. vacuum field amplitude at cavity centre is $E_{vac} = 0.79 \cdot 10^{-2}$ V/m. The cavity quality factor is $Q = 10^8$, corresponding to a photon damping time $2 \cdot 10^{-4}$ s. The average time spent by each atom in the mode is $2 \cdot 10^{-5}$ s. The relevant atomic energy levels are shown in fig. 1*b*). Before entering the cavity, the atoms are prepared by stepwise diode laser excitation in the $40P_{3/2}$ state and transferred by microwave excitation into the $39S_{1/2}$ state. The atomic flux is controlled by adjusting the last step diode laser intensity. The $39S_{1/2}$ hyperfine levels $F = 3$ and $F = 2$ (separated by a frequency interval $\Delta/2\pi = 320$ kHz, unresolved by the saturating microwave excitation process) are prepared with a statistical ratio 7/5. The cavity is exactly resonant with the upward transition coupling the $39S_{1/2}$, $F = 3$ sublevel to the $39P_{3/2}$ state. If one neglects the hyperfine coupling in this latter state (hyperfine constant $A_{39P_{3/2}}/2\pi = 7$ kHz), the $39S_{1/2} \to 39P_{3/2}$ transition is a doublet with frequencies $\omega_0/2\pi = 68.378\,562$ GHz ($39S_{1/2}$, $F = 3 \to 39P_{3/2}$) and $(\omega_0 + \Delta)/2\pi$ ($39S_{1/2}$, $F = 2 \to 39P_{3/2}$). The coupling of this multilevel atomic system to the cavity mode is *a priori* more complicated than the «dressing» of a two-level atom [1]. The situation remains simple however because the hyperfine coupling $A_{39P_{3/2}}$ is negligible compared to Ω_0. The atom-field coupling and the restriction of the hyperfine interaction to the $39S_{1/2}$ state can be simply diagonalized together [7]. Each of the twelve $39S_{1/2}$, $F = 3$ and $F = 2$ substates is coupled by the linearly polarized cavity field to a different substate of the $39P_{3/2}$ level. All these substates are mutually orthogonal. The atomic energy diagram thus consists in twelve sets of two-level systems, all independently coupled to the field with the same vacuum Rabi frequency at cavity centre $2\Omega_0/2\pi = 140$ kHz ([1]). Seven of these systems (corresponding to $39P_{1/2}$, $F = 3$) are resonant at ω_0 and five ($39S_{1/2}$, $F = 2$) at $\omega_0 + \Delta$. When the cavity mode is precisely tuned to ω_0, the $F = 2$ atoms are nonresonant enough so that their coupling to the mode can be in first approximation neglected and the experiment is then, at least qualitatively, analysable in terms of a two-level atom model.

In an experimental run, the cavity frequency is set to the value $\omega_0/2\pi$ (within ± 20 kHz) and a tuneable microwave field (angular frequency ν) is fed into the cavity through a wave guide connected to the atomic entrance hole. The probe is weak enough so that its perturbation on the atoms in the entrance wave guide is found to be negligible. The atoms are detected downstream by state selective field ionization. One thousand atomic events are sampled for each ν value. The fluxes n_s and n_p of atoms leaving the cavity in the $39S_{1/2}$ and $39P_{3/2}$ levels are deduced from the raw data by taking into account the decays of these levels during the atom flight time between the cavity and the detector. The transfer rate is defined as $\mathcal{T} = n_p/(n_s + n_p)$. A small constant background due to probe-independent spontaneous emission transfers between Rydberg states ([2]) is subtracted from the data. Figure 2 shows $\mathcal{T}$ *vs.* ν for two different atomic fluxes, decreasing from 2*a*) to 2*b*). The crosses are experimental and the line is a theoretical fit discussed below. Vacuum Rabi splittings between the two main peaks of 240 ($\pm$ 10) kHz and 170 ($\pm$ 10) kHz are clearly resolved in fig. 2*a*) and *b*), respectively.

The mean atomic flux is conveniently expressed in terms of the average number N_a of atoms in the cavity at a given time, which can be evaluated in several ways. A first value can be inferred from the Rabi splitting itself, using a qualitative model involving stationary two-

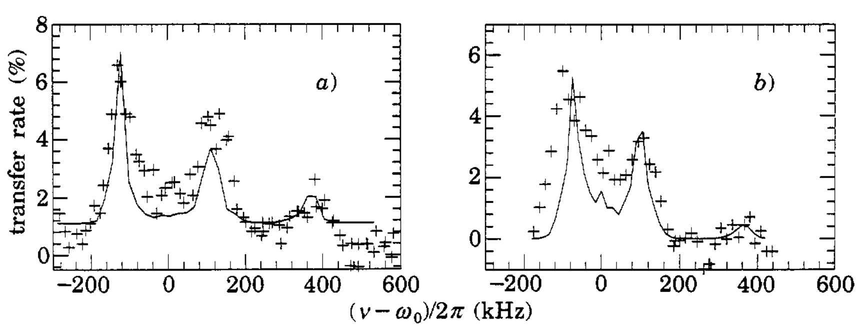

Fig. 2. – Vacuum Rabi splitting spectra corresponding to $N_a = 10$ (*a*)) and $N_a = 5$ (*b*)). The transfer rate $\mathcal{T} = n_p/(n_p + n_s)$ is plotted (in %) *vs.* $(\nu - \omega_0)/2\pi$ (in kHz). Crosses are experimental. Solid lines result from theoretical simulation. The «noise» on these lines is due to the random choices made in the simulation. Forty values of ν are sampled, for each of which a transfer rate is obtained by averaging 1000 (*a*)) or 500 (*b*)) atomic events.

([1]) $\Omega_0 = dE_{\mathrm{vac}}/\hbar$ with d being the matrix element of the z component of the atomic electric dipole between the electronic states $39S_{1/2}$, $m_J = 1/2$ and $39P_{3/2}$, $m_J = 1/2$ ($d = 5.9 \cdot 10^{-27}$ Coulomb meter).

([2]) A fraction of the atoms excited by the last diode laser step in the $40P_{3/2}$ level are not transferred in $39S_{1/2}$ during the microwave preparation stage. They subsequently cascade down by spontaneous emission to the $40S_{1/2}$ state which is detected in the same ionizing field as $39P_{3/2}$, and contribute to the background.

level atoms randomly distributed along the cavity axis. It can be shown [1] that the spectrum of this system exhibits a Rabi splitting identical to the one of a collection of atoms symmetrically coupled to the mode, with Ω_0 being replaced by $\Omega_0/\sqrt{2}$, which is the r.m.s. value of the coupling along the cavity axis. Furthermore, only $39S_{1/2}$, $F=3$ atoms are resonantly coupled to the cavity mode, so that the effective atom number to consider in a two-level model is $7N_a/12$. The splitting (in frequency units) is thus expected to be $(1/\sqrt{2})(2\Omega_0/2\pi)\sqrt{7N_a/12}$. This yields $N_a=10$ (± 1) and 5 (± 0.5) for fig. 2*a*) and *b*), respectively. A more direct, though less accurate estimate is made by measuring the total electron current resulting from Rydberg atoms ionized by an auxiliary electric field in the laser excitation zone. Each atom yields one electron and one ion which may subsequently produce secondary electrons by impinging on the surrounding metal surfaces. This method thus provides only an upper limit to the atomic flux, which corresponds to 15 ($\pm$ 5) atoms for spectrum 2*a*, compatible with the value quoted above.

We have recorded similar spectra for various atomic fluxes. Figure 3 shows a plot of the frequency splitting between the two main peaks as a function of $\sqrt{N_a}$, demonstrating the expected linear dependence. The smallest N_a value is determined by fitting the splitting to the two-level atom model prediction and the other atomic fluxes are deduced from the ratios of the atomic counting rates in the different spectra.

The qualitative model described above fails to explain two features of the spectra: the unbalance between the two main peaks and the existence of a weak third line around $\nu=\omega_0+\Delta$. This model overlooks the motion of the atoms, the fluctuations of the atom number in the cavity and most importantly, the nonresonant coupling of the $39S_{1/2}$, $F=2$ atoms to the cavity mode. We have performed a numerical simulation of the actual experimental situation in which the twelve sets of atomic levels are taken into account and the system evolution is explicitly analysed. The system is described by a wave function superposition of atom-cavity mode states whose evolution is ruled by a time-dependent atom field Hamiltonian reflecting the variation of the atom-field mode coupling during the cavity crossing time. The field relaxation is neglected. The coupling to the probe field is treated as a weak classical periodic perturbation of the cavity mode. Atoms are introduced in the cavity at random times, simulating the Poisson statistics of the atomic beam. The initial

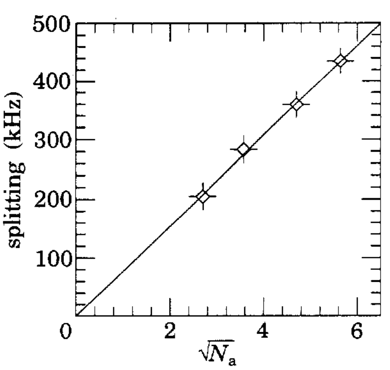

Fig. 3.

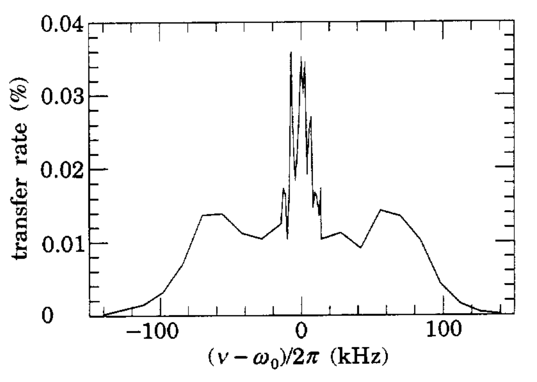

Fig. 4.

Fig. 3. – Vacuum Rabi splitting *vs.* square root of average atom number N_a.

Fig. 4. – Simulation of vacuum Rabi splitting spectrum for $N_a=1$. Each atom enters the cavity in the $39S_{1/2}$, $F=3$ state. Detection of 100 atoms is simulated for each ν value. The noise, larger than in fig. 2, is due to the smaller sampling. The central peak corresponds to the system response during time intervals when the cavity is empty.

populations of the $39S_{1/2}$, $F=2$ and $F=3$ states reflect their statistical weight (5/7). Atomic velocities are also randomly selected to reproduce the Maxwellian velocity distribution. The probabilities of detecting each atom exiting the cavity in the $39S_{1/2}$ and $39P_{3/2}$ states are computed and a random number generator is used to simulate the outcome of each atom detection process. After each of these processes, the atom field system is «reduced» into a state of the remaining $(N-1)$ atom + field system which depends upon the outcome of the detection, according to the usual postulate of a quantum measurement. The transfer rate is computed for each ν value by counting atoms in levels $39S_{1/2}$ and $39P_{3/2}$ during a time interval corresponding to 100 atom-cavity interaction time (statistics performed on $100N_a$ atoms). The transfer rate being at low intensity proportional to the probe power, the simulation is performed with a very weak probe, which minimizes the number of excited atom-field states in the calculation and saves computer time. The results are then normalized to fit the vertical experimental scale. The mean atom number N_a is the only adjustable parameter left. The best fits in fig. 2*a*) and *b*) correspond to $N_a=10$ and 5, in excellent agreement with the qualitative two-level atom model estimate. The frequencies and relative intensities of the two main peaks and small third line are well accounted for. A change of N_a by one unit results in a noticeable discrepancy between the experiment and the simulation, which is an indication of the sensitivity of our atom number calibration. Qualitatively, the third peak corresponds to the $39S_{1/2}$, $F=2 \rightarrow 39P_{3/2}$ transition induced by the small probe field penetrating in the atom-perturbed cavity at a frequency close to $\nu=\omega_0+\Delta$. The unbalance between the two main lines can be attributed to coupling between atoms via their common interaction with the cavity vacuum. This results in a coupling of the two sets of atomic oscillators which perturbs their normal-mode amplitudes.

The normal-mode splitting linewidth is partly determined by transit time effects. Since the atom-cavity interaction time is much shorter than the photon damping time, field relaxation has a negligible contribution, which justifies leaving it out of the simulations. Another contribution to the linewidth is expected from the fluctuation of the number of atoms in the cavity around N_a, which causes related fluctuations in the splitting. These effects are included in the theory which predicts 60 kHz linewidths. The recorded spectra exhibit larger linewidths of the order of 120 kHz. We attribute the difference to $a \approx 100$ kHz Stark broadening of the atomic levels due to stray electric fields in the cavity of the order of 100 mV/cm (contact potentials, path effects). These fields are even larger in the wave guide where the atoms are prepared before they enter the cavity (they pass within 0.5 mm of the metal surfaces). We have not attempted to include the Stark effect in our simulations because we do not know the exact geometry of the fields.

In this experiment, we have resolved the vacuum Rabi splitting doublet for atom numbers an order of magnitude smaller than previously published ($N_a=40$ in [5]). Our minimum number $N_a=5$ corresponds in fact to only 3 atoms on average resonant with the cavity mode. The splitting is not clearly resolved at lower fluxes, but spectra for $N_a=2$ to 3 (*i.e* $7N_a/12=1$ or 2) exhibit lineshape modifications which can be attributed to the fluctuations of the index of refraction of a resonant medium made of one or two atoms! If one notes that Kimble *et al.* have recently reported a yet unpublished normal splitting with 2 atoms in an optical Fabry-Perot [8], it appears that observation on a single atom is not far away. In our experiments, the Stark broadening is a major source of difficulty and becomes of the order of the splitting when $N_a=1$. We plan to resume the experiment with a larger cavity of a different design where we expect the stray fields to be less a problem. It should also be possible to optically pump all the atoms in the $39S_{1/2}$, $F=3$ level, thus realizing a true two-level system for which the splitting effect is symmetrical (Stark effect rendered optical pumping impossible in our experiment by scrambling the populations between the $F=3$ and $F=2$ states in the cavity entrance wave guide). Our numerical simulations predict that the one-atom splitting should then become observable. Figure 4 shows the theoretical spectrum in the case of a beam of atoms prepared in the $39S_{1/2}$, $F = 3$ state, with a flux corresponding to an average value $N_a = 1$. The probability of having one or zero atom in the cavity is then the same (0.37), with 2 atoms in the cavity 18% of the time and 3 atoms only 6% of the time. Two symmetrical peaks separated by 120 kHz are expected, with a third central line at $\nu = \omega_0$. This extra feature corresponds to the free cavity response to the probe field during the time intervals when the cavity is empty. The energy which is then fed into the cavity is absorbed by the atoms which subsequently interact with the mode and is carried away in the form of atomic excitation and recorded by the field ionization detection. This central feature disappears when N_a is of the order of two or more, since the cavity is then practically never empty.

We have considered in this letter the resonant atom-cavity coupling case. When the cavity mode frequency ω differs from ω_0 by an increasing amount δ, the two lines of the single-atom vacuum Rabi doublet continuously evolve, at the $|\delta| \gg \Omega_0$ limit, into a «cavity line» at frequency $\omega + \Omega_0^2/\delta$ and an «atomic line» at frequency $\omega_0 - \Omega_0^2/\delta$ [1]. The quantity Ω_0^2/δ thus appears as a «pulling» of the cavity mode produced by a single nonresonant atom or as a «light shift» of the atomic transition frequency induced by the cavity vacuum. If instead of being initially empty, the cavity contains η photons, this light shift becomes $n\Omega_0^2/\delta$ and Ω_0^2/δ can also be interpreted as the atomic frequency shift «per cavity photon». Detection of these shifts can be turned into a very efficient way of quantum nondemolition measurement for small microwave field photon numbers [6]. By investigating radiative atom-cavity shifts in the resonant case, the experiment reported here is an encouraging step towards these challenging measurements. It shows that resonant shifts in the 100 kHz range can realistically be observed in single Rydberg atom cavity QED experiments. With a $\delta/2\pi = 500$ kHz detuning, nonresonant shifts of about 10 kHz are expected for atoms at cavity centre. This order of magnitude is well within reach of high-resolution microwave spectroscopy techniques [6].

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