One-Atom Maser

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The exchange of photons between single Rydberg atoms and a single mode of a superconducting cavity with a quality factor $Q = 8 \times 10^8$ at 2 K was observed. Signals could still be detected with an average number of only 0.06 atom simultaneously in the cavity. With one Rydberg atom the linewidth of the maser transition at about 21 GHz was power broadened and at higher densities asymmetry of the transition was observed, which is ascribed to an ac Stark effect.

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The most basic problem of radiation-matter coupling is the interaction of a single two-level atom with a single-mode electromagnetic field. This problem received a great deal of attention shortly after the maser was invented.¹ At that time the problem was of purely academic interest, since in the experiments it was always necessary to have a large number of atoms and photons. This was due to the fact that it was impossible to detect small amounts of atoms, and that the small size of the transition matrix elements resulted in atom-field coupling times much longer than other characteristic times of the system, such as atomic relaxation or the interaction time with the field.

This situation completely changed when the advent of frequency-tunable lasers allowed the study of the highly excited Rydberg states of atoms, for three reasons. First, these states are very strongly coupled to the radiation field. Second, the transitions to neighboring levels are in the region of millimeter waves, which allows one to build cavities with low-order modes being sufficiently large to ensure rather long interaction times. Finally, the Rydberg atoms have long spontaneous lifetimes, so that only the coupling with the selected cavity mode is important. The theories of the one-atom, one-mode problem predict a few interesting and basic effects that can now be studied experimentally. They result from modifications of the boundary conditions for the vacuum field around the atom, due to the presence of the cavity walls. The following are examples: The spontaneous emission of a single atom in the cavity is expected to be drastically modified as compared with its behavior in free space; spontaneous emission is enhanced in a resonant cavi ty^2 and suppressed if the cavity is off resonant.³ Furthermore, if the cavity finesse is very high, an oscillatory energy exchange between a single atom and the cavity mode is expected to occur.¹ If a field is initially present in the resonator, being produced either by a thermal or a coherent source, various types of

dynamics are predicted, such as, e.g., the disappearand revival of the Rabi nutation.⁴ (For a review of these phenomena see Haroche.⁵)

Enhancement of the spontaneous emission rate of a single atom prepared in a tuned cavity has already been observed.⁶ However, in this experiment the oscillatory energy exchange between the atom and field could not be observed since the losses in the cavity used were still too large. This process has now been observed in the experiment described in the following.

The Rydberg maser experiment employs an atomic beam to ensure collision-free conditions for the highly excited Rydberg states. A diagram of the vacuum chamber with the atomic-beam arrangement and microwave cavity is shown in Fig. 1. These parts are mounted inside a helium-bath cryostat. Rubidium atoms were used for the experiment. The atomicbeam oven is carefully shielded from the cavity by cooper plates cooled by water, liquid nitrogen, and liquid helium. The beam passes through small apertures into the liquid-helium-cooled part of the apparatus. There the atoms are pumped by the laser radiation to the upper maser level and enter the cavity. Behind the cavity the atoms are monitored by field ionization and subsequent detection of the ejected electrons in a channeltron electron multiplier.

The Rydberg states were excited with the frequency-doubled light of a commercial cw ring dye laser (Coherent CR 699-21). The second harmonic was generated by means of a temperature-stabilized amonium dihydrogen arsenate crystal. In a single pass, about 60 μ W of ultraviolet radiation at 297 nm was obtained. This laser power was sufficient to excite up to 30 000 atoms/s in the upper maser level $63p_{3/2}$ of the more abundant (70%) ⁸⁵Rb isotope. The wavelength of the laser light is measured by means of an air-track lambdameter. An additional check of the wavelength was performed by observation of the well-known iodine absorption spectrum. For this purpose the light



FIG. 1. Vacuum chamber with the atomic-beam arrangement and the microwave cavity. The upper part is cooled to liquid-helium temperature.

of the dye laser is transmitted through an iodine cell and the intensity is monitored by a photodiode. Finestructure splitting between $63p_{3/2}$ and $63p_{1/2}$ of ⁸⁵Rb amounts to 396 MHz. It is, therefore, no problem to excite a single fine-structure level of ⁸⁵Rb with the narrow-band ultraviolet radiation ($\Delta \nu \approx 2$ MHz).

The polarization of the incident ultraviolet laser light is perpendicular to the atomic beam and parallel to the direction of the electric field in the cavity. In this way $\Delta m_j = 0$ are the only allowed transitions. The atomic beam passes through the cylindrical cavity along its axis, where only the TE_{1np} and TM_{1np} modes posses a nonvanishing transversal-electric field. For our experiment the TE₁₂₁ mode was used. The variation of the field strength of this mode over the cross section of the atomic beam (0.5 mm) is less than 2%. The frequency of the maser transition [21 506.51(5) MHz] was determined in separate double-resonance experiments. This was necessary since the tuning range of the cavity is very small, and required that the cavity resonance be nearly equal to the atomic resonance.

The TE₁₂₁ mode has a plane-field distribution and is doubly degenerate in an ideal cylindrical cavity. The degeneracy is removed by a slight deformation of the circular cross section into an oval shape, which then determines the direction of polarization of the field mode. The deformation is achieved by our squeezing the cylinder both with a screw and a piezoelectric transducer crystal ($\Delta l \approx 4 \,\mu$ m/1500 V at 2 K) for fine tuning. This causes the one degenerate resonance to be shifted towards higher frequencies, whereas that for orthogonal polarization shifts to lower values. The field polarization important for our experiment is coupled to an external waveguide so that the cavity performance can be tested. The upper frequency branch used in our experiment can be mechanically tuned by about 15 MHz; the piezoelectric drive can sweep the frequency by 0.5 MHz/1500 V.

The cylindrical cavity has a diameter of 24.7 mm and a length of 24 mm. It was manufactured from pure niobium rods (99.9%, Kawecki Berylco, Inc.) in two parts: a cylindrical jar and a cover with the waveguide coupling. Constructing the cavity in this fashion has a negligible effect on the cavity Q. Both parts were recrystallized several hours in an ultrahigh vacuum oven and then chemically polished ($\approx 100 \ \mu m$). After assembly and electron-beam welding, the cavity was again chemically polished and then annealed for 8 h in an ultrahigh vacuum oven at 2000 °C. The cavity was enclosed in a Mumetal shield to reduce the influence of ambient magnetic fields on the O value due to frozen-in flux. The properties of the high-frequency superconductivity were thoroughly investigated in a bath cryostat, where the cavity was cooled by direct contact with liquid helium. For these measurements a second waveguide coupling was attached to the jar, and the cavity was evacuated. The surface resistance was obtained from the Q value (1089 Ω/Q), which was determined from the decay time of the stored energy (decrement method). The comparison with BCS theory gave a reduced energy gap of $\Delta/kT_c = 1.854(6)$ in the temperature range between 1.43 and 4.27 K for the TE_{121} mode.

The temperature of the cavity in the atomic-beam arrangement was measured by means of a germanium resistor. It could be varied from 4.3 to 2.0 K, corresponding to Q factors of 1.7×10^7 and 8×10^8 , respectively. To shield the 300-K thermal radiation of the test equipment, a cooled flap with a temperature of less than 10 K was inserted into the waveguide at the upper end of the cavity. The thermal-background field inside the niobium cavity is therefore essentially determined by the temperature of the walls.

The average number of photons of the blackbody radiation is given by $n_{bl} = [\exp(h\nu/kT) - 1]^{-1}$, being about $n_{bl} = 3.8$ at 4.3 K and 1.5 at 2 K. Field ionization is the standard technique for the detection of Rydberg atoms. The field strength necessary for ionization scales as $(n^*)^{-4}$. It is therefore possible to distinguish between Rydberg states belonging to different main quantum numbers. If the maximum field strength is chosen properly ($\cong 20$ V/cm), atoms in the 63*p* state are ionized and detected by our counting the electrons with a channeltron electron multiplier. At the same field strength, atoms in the neighboring 61*d* level are ionized with a smaller probability ($\cong 15\%$ of the 63p state).

Transitions from the initially prepared $63p_{3/2}$ state to the $61d_{3/2}$ level are thus detected by reduction of the electron count rate. The detector is placed 63 mm downstream from the point where the atoms are excited. Considering the lifetime of the $63p_{3/2}$ level, one finds that $\approx 70\%$ of the excited atoms reach the detector.⁷

To demonstrate maser operation, the cavity was tuned over the $63p_{3/2}$ - $61d_{3/2}$ transition by changing the voltage of the piezoelectric transducer; the field-ionization signal was recorded simultaneously. The signal transients could be averaged by means of the PDP 11/10 computer. About 10 to 20 transients were averaged for most of the measurements. The signal obtained with a cavity temperature of 4.3 K is shown in Fig. 2. The reduction of the count rate on the maser resonance was about 28%. Up to particle fluxes of 22×10^3 atoms/s no change in the shape and position of the resonance could be observed.

In the case of the measurements at a cavity temperature of 2 K (Fig. 3), a reduction of the $63p_{3/2}$ signal could be clearly seen for fluxes as small as 800 atoms/s. An increase of the flux causes power broadening and finally asymmetry and a small shift. The shift has to be attributed to the ac Stark effect, caused predominantly by virtual transitions to the $61d_{5/2}$ level, which is only 50 MHz away from the maser transition. The fact that the field-ionization signal at resonance is independent of the particle flux (between 800 and 22×10^3 atoms/s) indicates that the transition is saturated. This fact and the observed power broadening show that there is a multiple exchange of photons between the Rydberg atoms and the cavity field.

With an average transit time of the Rydberg atoms



FIG. 2. Maser resonance at a cavity temperature of 4.3 K. Flux of the Rydberg atoms in the upper maser level N = 2000/s.

through the cavity of 80 μ s, one calculates for a flux of 800 atoms/s a probability of 0.06 of finding a Rydberg atom in the cavity. According to Poissonian statistics this implies that more than 99% of the events are single atom. This clearly demonstrates that single atoms are able to maintain continuous oscillation of the cavity. Since the transition is saturated, half of the atoms initially excited in the $63p_{3/2}$ state leave the cavity in the lower $61d_{3/2}$ maser level. The decay to other levels can be neglected for the average transit time of 80 μ s. The energy radiated by these atoms is stored in the cavity field for the characteristic cavity decay time, increasing the average field strength.

The average number of photons left in the cavity by the Rydberg atoms is given by

$$n = \tau_d N/2,$$

where τ_d is the characteristic decay time of the cavity and N the number of Rydberg atoms in the upper maser level entering the cavity per unit time. For the highest particle flux used in our experiment, $N=22 \times 10^3$ atoms/s, one calculates that $n \approx 55$ photons at 2 K ($\tau_d \approx 5$ ms). At 4.3 K ($\tau_d \approx 0.13$ ms) there is an average number of $n \approx 1.4$ photons. This number is smaller than the average number of blackbody photons, about 4 at 4.3 K. At a temperature of 2 K the average number of blackbody photons is $n_{bl} \approx 1.5$. In the case of $N \approx 800$ atoms/s one obtains $n \approx 2$, which means that the energy of the radiation generated by the Rydberg atoms in the cavity is about the same size as that of the blackbody radiation.

When the squares of the half-widths $\Delta \nu$ of the signal curves are plotted versus the Rydberg atom flux, a straight line is obtained as expected (Fig. 4). This line intersects the $(\Delta \nu)^2$ axis at a finite value, from which the number of blackbody photons originally in the cavity can be evaluated. The result (3 ± 1) is in reason-



FIG. 3. Maser resonance at a cavity temperature of 2 K.



FIG. 4. Squares of the half-widths of the maser resonances vs atomic flux. From the intersection of the straight line with the $(\Delta \nu)^2$ axis, the number of blackbody photons in the cavity can be evaluated. This was done with use of the Rabi frequency $\Omega = 43$ kHz, which was evaluated from the maser data at 4.3 K.

able agreement with the value given above. It follows that as the atomic flux decreases the thermal radiation becomes the dominant part of the field.

The coupling constant between the atom and radiation is big enough to allow a multiple exchange of photons between the cavity mode and a single Rydberg atom to occur under these conditions. It follows that under the conditions shown in Fig. 3 the atom performs on the average of about 5 to 20 Rabi periods when passing through the cavity.

Because of the velocity distribution of the atoms it is not possible to observe the Rabi nutation direct. At present a Fizeau-type velocity selector is inserted between the atomic beam oven and the cavity. This will enable us to observe the Rabi nutation of the atoms directly, because then we will have a fixed interaction time of the atom with the cavity field. Changing the selected velocity leads to a different interaction time and leaves the atom in another phase of the Rabi cycle when it arrives at the detector. In this way more-detailed studies of the atom-field interaction than in the present experiment will be possible. In particular, there is a good chance that experimental observation of the predicted disappearance and revival of the Rabi nutation will also become possible.

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