Realization of a Two-Photon Maser Oscillator

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We have built the first quantum oscillator working on two-photon stimulated emission of radiation. It oscillates continuously on a degenerate two-photon transition between the levels $40S$ and $39S$ of rubidium atoms. The oscillation is sustained in a $Q \simeq 10^8$ niobium superconducting cavity at 68.41587 GHz. At threshold, this new maser system operates with only a few atoms and a few tens of photons at a time in the cavity.

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Quantum amplifiers or oscillators operating on a two-photon atomic transition were proposed more than twenty years ago[1] and the theory of these systems has been studied in many papers since then.[2,3] They are expected to differ from ordinary one-photon lasers and masers in many respects, such as the nature of the phase transition corresponding to the onset of emission, the statistics of the radiation field, and their multistable and hysteresis behavior. Experimental study of these systems is highly desirable and has already prompted several attempts to build a working device. Evidence for two-photon stimulated emission has been reported.[4] Emission of light pulses at frequency ω_1 stimulated by the injection of a pulsed beam at frequency ω_2 (where $\omega_1+\omega_2$ is equal to the atomic transition frequency) has also been observed.[5] However, there has been no success so far in realizing a genuine two-photon oscillator in which an inverted atomic medium placed in a cavity radiates on a two-photon transition without assistance from an external radiation field. Usually the gain on a two-photon transition is exceedingly small and the atomic densities required to achieve oscillation are so large that unwanted competing effects (collisions, nonlinear processes such as multiple-wave mixing, stimulated Raman effect) are dominant. We have recently proposed[6] to overcome these difficulties by employing Rydberg atoms as the active medium in a high-Q superconducing microwave cavity. In this Letter, we report the realization of this system and the continuous-wave operation of the first two-photon quantum oscillator.

The advantages of our using transitions between neighboring Rydberg states for maser studies are manifold.[7] The electric dipole matrix elements are exceedingly large; these transitions correspond to millimeter wavelengths for which it is possible to realize high-Q low-order-mode cavities, very efficiently coupled to the atomic dipoles. These properties have made it possible to build masers operating on one-photon transitions with only one atom at a time in the cavity.[8] An additional feature makes alkali-metal-atom Rydberg states especially convenient for two-photon maser studies. The two-photon transition can be chosen so that there is an intermediate level i nearly halfway between the initial level e and final level f, thus greatly enhancing the transition amplitude. This remarkable property has been analyzed in detail in Ref. 6 and the theory of the Rydberg-atom two-photon maser operation expounded by Brune and co-workers.[6,9]

The level scheme of our two-photon oscillator is shown in Fig. 1. Maser action occurs between the $40S_{1/2}$ state (e) and the $39S_{1/2}$ state (f) of ^{85}Rb (energies E_e and E_f, respectively). The atoms emit in a high-Q cavity tuned at frequency $\nu=(E_e-E_f)/2h=68.41587$ GHz (two-photon degenerate transition). Levels e and f have the same parity. The opposite-parity intermediate level $39P_{3/2}$ (i) is connected to both e and f states by electric-dipole–allowed transitions. The dipole matrix elements D_{ei} and D_{if} are nearly equal and very large (1500 a.u.). The level i is detuned by the amount $h\Delta=E_i-(E_e+E_f)/2$ from the average of the energies of the e and f states ($\Delta/2\pi=-39$ MHz only). The atom-field coupling is described by the elementary one-photon Rabi angular frequency $\Omega=D_{ei}\mathcal{E}_0/\hbar \simeq D_{if}\mathcal{E}_0/\hbar$, where $\mathcal{E}_0=[h\nu/2\epsilon_0 V]^{1/2}$ is the field per photon in the cavity (effective volume $V=70$ mm^3, $\Omega=7\times10^5$ s^{-1}). When the cavity contains N photons, the populations of levels e and f undergo a second-order Rabi precession at the angular frequency

$$\Omega_{ef}(N)=\Omega^2(2N+3)/\Delta=A+BN. \qquad (1)$$

The frequencies A and B, inversely proportional to Δ, are unusually large in this system ($A=6\times10^3$ s^{-1} and $B=4\times10^3$ s^{-1}). The A term corresponds to the two-photon spontaneous-emission precession in the cavity mode and the BN term to the two-photon stimulated pre-

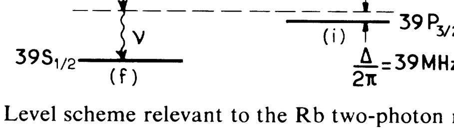

FIG. 1. Level scheme relevant to the Rb two-photon maser.

cession at a frequency proportional to the field intensity in the cavity. Equation (1) is based on a three-level approximation of the two-photon process, amply justified by the very small value of $\Delta/2\pi\nu$ ($\simeq 6\times10^{-4}$).

The atoms excited in level e cross the cavity at thermal velocity ($v \simeq 300$ m/s), in an average time $t_{\rm int} \simeq 25$ μs. The average atomic flux is $t_{\rm at}^{-1}$. The atoms release pairs of photons in the cavity, which are damped away within the time $t_{\rm cav} = Q/\omega$ ($t_{\rm cav} = 2.3\times10^{-4}$ s for $Q \simeq 10^8$). In the simplest model, the average number $\bar{N}$ of photons around threshold for this system must satisfy the following equations[6]:

$$\Omega_{ef}(\bar{N})t_{\rm int} \simeq \pi, \qquad (2)$$

$$\bar{N} = 2t_{\rm cav}/t_{\rm at}. \qquad (3)$$

Equation (2) means that each atom has a large probability of releasing its energy into the field and Eq. (3) implies that the atomic flux is able to maintain the field in the cavity in spite of the losses. Combining Eqs. (1), (2), and (3), we get a threshold flux

$$(1/t_{\rm at})_{\rm th} = \pi/2Bt_{\rm int}t_{\rm cav} \simeq 7\times10^4\ {\rm s}^{-1}$$

and $\bar{N} = 32$. A more realistic theory also takes into account velocity dispersion, atomic losses due to spontaneous emission towards lower states during $t_{\rm int}$, and the field distribution in the cavity. It yields the somewhat larger threshold $(1/t_{\rm at})_{\rm th} = 1.7\times10^5$ s^{-1} and $\bar{N} = 40$. For such low atomic fluxes and small radiation energies, all other competing effects remain negligible. In particular, the one-photon Rabi frequency $\Omega(\bar{N})^{1/2}/2\pi$ remains small compared to $\Delta/2\pi$ so that one-photon transitions are not power broadened enough to come in resonance with the cavity mode.

A more complete description of this maser shows that above threshold it generally presents several metastable operating points, among which is the $\bar{N} = 0$ state.[3,9] If the two-photon maser operates with very large photon numbers (classical limit), theory indicates that the system cannot start without being triggered away from the $\bar{N} = 0$ state by an external field.[6] When the maser is in a microscopic regime (very small atom and photon numbers), quantum fluctuations [in particular the two-photon spontaneous-emission process described by the A term in Eq. (1)] are shown to make the $\bar{N} = 0$ point unstable and the maser field is expected to start spontaneously after a delay which can vary from a few times $t_{\rm cav}$ to infinity.[9]

The Rb-atom excitation scheme and the experimental setup are shown in Fig. 2. A beam of rubidium atoms is excited into the $40S_{1/2}$ state by a four-step continuous-wave process. Three frequency-stabilized diode-laser beams carry the excitation to the $40P_{3/2}$ level via the $5P_{3/2}$ and $5D_{5/2}$ states[10] ($\lambda_1 = 7802.4$ Å, $\lambda_2 = 7759.8$ Å, $\lambda_3 = 12644.1$ Å). The last $40P_{3/2}$ to $40S_{1/2}$ excitation at $\nu_1 = 62.99$ GHz is achieved just before the atoms enter the cavity with microwaves produced by an X-band klystron, frequency multiplied with a Sielux harmonic generator.[11] It is important to notice that the microwaves at ν_1 do not enter in the cavity tuned at the different frequency ν. In this way we produce a 1.3-mm^2, 1.3-mrad angular-aperture beam of Rb atoms in the $40S_{1/2}$ state,[12], with atomic fluxes up to $t_{\rm at}^{-1} = 10^7$ s^{-1}. The excited atoms enter the liquid-He–cooled cylindrical Nb cavity (length, 7.5 mm; diam 7.7 mm) operating in the TE_{121} mode. The cavity (machined, electron-beam welded, and annealed at CERN[13]) has $Q \simeq 10^8$ at 1.7 K (decreasing to 3×10^7 at 2.5 K). The average number of thermal photons in the mode at 1.7 K is 0.17; blackbody effects are negligible.

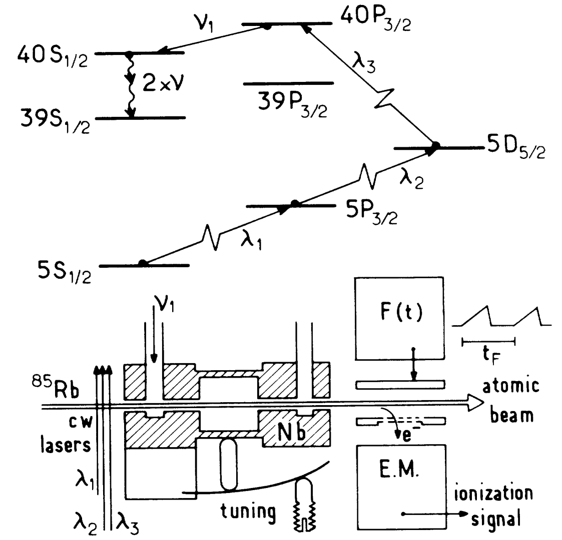

FIG. 2. Excitation scheme for the Rb atoms (top) and sketch of the two-photon maser setup (bottom).

Fine tuning of the cavity frequency to the two-photon resonance line is one of the critical aspects of the experiment. The cavity is initially resonant about 50 MHz below the two-photon frequency—68.41587 GHz—which has been determined with an accuracy of ± 10 kHz in a preliminary two-photon-spectroscopy experiment performed in our atomic beam. Tuning of the cavity is achieved by mechanical deformation. An elastic blade converts the turn of a screw into a force pushing the cavity wall (see Fig. 2). Deformations produced at low temperatures are elastic, so that reversible tuning across resonance is possible. The position and width ($\simeq 800$ Hz at 1.7 K) of the cavity resonance are monitored during tuning by measurement of its transmission versus frequency with a millimeter-wave scalar network analyzer (Sielux MSNA27-125) with a 70-dB dynamic range.[11] This analyzer is frequency locked to a Microwave System MOS3106 frequency stabilizer providing an overall resolution of 200 Hz (this system is

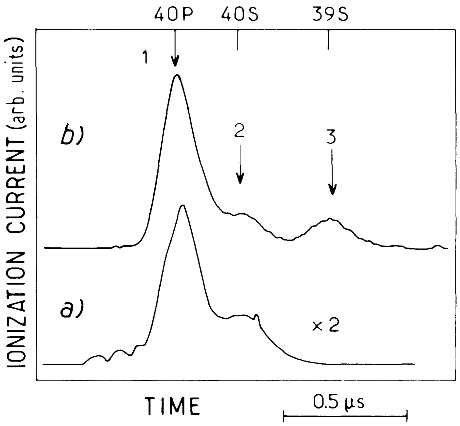

FIG. 3. Time-resolved atomic ionization signals (trace a) below threshold and (trace b) above threshold for two-photon maser emission. The arrows indicate the times at which levels $40P$, $40S$, and $39S$ ionize in the electric field ramp. Appearance of peak 3 in trace (b) is evidence for maser action. Detection sensitivity is twice as large in trace (a) as in trace (b).

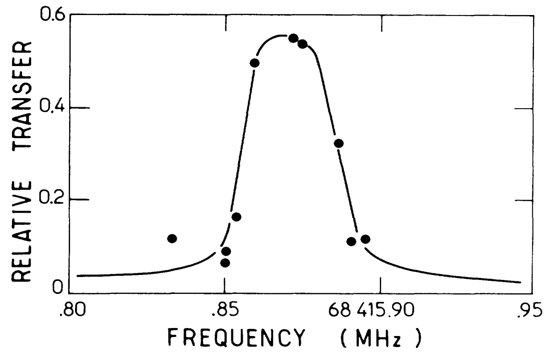

FIG. 4. Maser signal vs cavity frequency. For visual convenience, the experimental points have been connected by a smooth line.

switched off when atoms cross the cavity). In this way, the cavity can be tuned to any desired frequency in a 150-MHz range with a ± 1 kHz accuracy and drifts by less than 2 kHz per hour.

The two-photon emission is monitored by measurement of the level populations of the atoms leaving the cavity, with use of the field-ionization method. The atoms cross a condenser in which a train of linearly rising electric field pulses $F(t)$ is applied. The pulse repetition rate $t_F^{-1}=7$ kHz is such that, on average, one atom out of seven is ionized. The field in each pulse reaches at different times the ionization threshold for the various levels involved in the process (the more excited the level, the smaller the electric field). The resulting electrons are detected by an electron multiplier. In this way, a time-resolved spectrum of the atomic populations is performed at the electric field pulse rate. A computer-controlled fast-transient digitizer (Sielux FTD) samples these signals and displays in real time (each 200 ms) a 1400-pulse average on the computer screen. Figure 3(a) shows the ionization signal obtained with a $Q=8\times10^7$ resonant cavity, for a small atomic flux. Peaks labeled 1 and 2 correspond respectively to the $40P_{3/2}$ and $40S_{1/2}$ levels prepared by the diode-laser–microwave excitation process.[14] When the atomic flux is increased, the signal shape suddenly changes to the typical signal shown in Fig. 3(b). Peak 3 corresponds to the $39S_{1/2}$ state, whose population is evidence of the two-photon maser action occurring in the cavity. Figures 3(a) and 3(b), respectively, correspond to $\simeq 80$ and $\simeq 200$ atoms detected per second in the $40S_{1/2}$ and $39S_{1/2}$ levels. We estimate that only a fraction $(1\pm0.5)\times10^{-3}$ of atoms entering the cavity in the $40S$ state is detected (losses include spontaneous emission towards lower-lying states, detection duty cycle, condenser grid transmission, and electron multiplier efficiency). Figures 3(a) and 3(b) thus correspond to $(8\pm4)\times10^4$ and $(2\pm1)\times10^5$ atoms per second entering the cavity, respectively, in good agreement with the predicted threshold.

The effect of cavity tuning is shown in Fig. 4. We have plotted the maser transfer rate (final population of the $39S_{1/2}$ state divided by the total population of the $40S_{1/2}$ and $39S_{1/2}$ levels) as a function of the cavity frequency, for an average atomic flux $t_{at}^{-1}=(2\pm1)\times10^5$ s^{-1}. The tuning range ($\simeq 50$ kHz) is centered on the independently determined two-photon transition frequency and is three orders of magnitude smaller than the frequency detuning of the $40S+39P$ one-photon transition (39 MHz). The small tuning range of this maser around frequency ν is a clear evidence of a true two-photon maser operation. In order to make an even more stringent test, ruling out the possibility of a $40S_{1/2}\to 39P_{3/2}\to 39S_{1/2}$ cascade, we have also checked the population of the intermediate level i ($39P_{3/2}$). This level ionizes in nearly the same field as the $40S_{1/2}$ state, so that it cannot be directly monitored. We have applied, on the downstream side of the cavity, a microwave field at the frequency of the $39P_{3/2}\to 37D_{5/2}$ transition (99.3 GHz) and checked that the ionization spectra corresponding to an operating maser were not changed. This is a clear indication that the $39P_{3/2}$ state is not populated at all and is thus only virtually involved in the two-photon maser operation.

When the two-photon–maser population inversion is decreased down to threshold, theory predicts that the start-up time diverges very rapidly.[9] Under specific conditions (maser oscillating at the edge of the cavity tuning range, or maser operating in a $Q\simeq3\times10^7$ cavity at 2.5 K), we have observed very long start-up times which might be a manifestation of this phenomenon. Switching on the laser pumping suddenly and slightly above threshold, we have observed that the ionization peak 3, initially lacking, abruptly appears after a delay of a few seconds and remains stable thereafter. More detailed investigation of this very striking transient behavior is underway. Other interesting effects we are presently studying include dynamical Stark shifts of the two-photon transition, hysteresis and multistable behavior of maser operation, and statistical fluctuations of atomic observables.

Let us stress in conclusion some orders of magnitude. The average number of atoms in the cavity, t_{int}/t_{at}, is about five around threshold. A moderate and feasible increase of Q would make the system operate in the true micromaser regime (one atom or less in the cavity), a remarkable feature for an oscillator operating on a two-photon effect. The total energy stored in the cavity is 10^{-2} eV around threshold, 2×10^{-1} eV for the strongest observed signal ($\bar{N}$ varying from 40 around threshold to $\simeq 800$). The power radiation in the output coupling waveguide is estimated to be 10^{-18} to 2×10^{-17} W. Direct detection of this very small microwave signal would provide very interesting complementary information on this novel quantum optics system.

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[12]We neglected so far the hyperfine structure (S states are split into two sublevels with angular momenta $F=2$ and 3). The stepwise excitation is tuned to excite mainly the $40S_{1/2}$, $F=3$ sublevel, which is coupled only to $39S_{1/2}$, $F=3$ ($\Delta F=0$ two-photon selection rule). e and f are thus unambiguously defined.

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[14]The saturating microwave field at frequency ν_1 nearly equalizes the $40P$ and $40S$ state populations. In Fig. 3, peak 2 is smaller than peak 1 because the $40S$ lifetime is shorter than the $40P$ one; more $40S$ than $40P$ atoms are lost by natural decay in the time interval between excitation and detection.