Generating and probing a two-photon Fock state with a single atom in a cavity

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Abstract

A two-photon Fock state is prepared in a cavity sustaining a “source mode” and a “target mode”, with a single circular Rydberg atom. In a third-order Raman process, the atom emits a photon in the target while scattering one photon from the source into the target. The final two-photon state is probed by measuring by Ramsey interferometry the cavity light shifts induced by the target field on the same atom. Extensions to other multi-photon processes and to a new type of micromaser are briefly discussed.

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The generation of non-classical states of light has been the subject of an intense theoretical and experimental activity since the first observations of squeezed states of light [1]. The study of these states provides a fundamental understanding of quantum fluctuations and opens the way for new schemes of communication or imaging beating the standard quantum noise limit. Entangled states produced in down-conversion processes have been widely used to test fundamental quantum features such as non-locality [2] or to realize quantum information transmission schemes (cryptography [3] or teleportation [4]). Quantum superpositions of fields with different classical parameters have been used to explore the quantum/classical boundary and the decoherence phenomenon [5].

In this context, Fock or photon number states are particularly interesting. They exhibit no intensity fluctuations and a complete phase indetermination. Their Wigner function presents negativities which reveal clearly their non-classical nature [6,7]. Many studies have been devoted to their production and characterization. Single-photon Fock states are easily prepared from a pair of entangled photons by the detection of one of the members of the pair [8]. They can also be prepared “on demand” by controlling the emission of a single radiator: molecule [9], colored center [10] or quantum dot [11]. These single-photon states are useful for quantum information transmission schemes [3]. Single-photon sources could also be an essential resource in an all-optical quantum information processing device [12].

Various schemes of Fock state preparation have been proposed in the context of Cavity Quantum Electrodynamics (CQED) [13], in which atoms interact one at a time with a high \( Q \) resonator. A one-photon Fock state has been produced in this way by a \( \pi \) quantum Rabi pulse in a microwave cavity [14] or by an adiabatic passage sequence in an optical cavity [15]. With a train of atoms, arbitrary multiphoton Fock states can in principle be generated in a cavity by post-selecting the events in which each atom has emitted one photon. Two-photon states have been generated in this way [16]. The operation of c.w. micromasers can also lead to Fock states preparation. The micromaser field evolves into a number state, provided the atomic velocity is conveniently tuned (trapping states [17]). A one-photon Fock state has been generated and probed in this way [18]. In all these CQED processes, at least one
atom per produced photon is required. Schemes more economical in atomic resources have been proposed [19,20] but never implemented so far.

We report in this Letter the production of a two-photon Fock state by a single atom in a high-$Q$ cavity. The scheme involves the non-resonant interaction of the atom with two non-degenerate cavity modes. The atom, in a single non-linear process, emits one photon in the initially empty target mode, while transferring another photon from the source mode (containing initially a small field) into the target. This third-order Raman process, resonant for proper atom-cavity mode detunings, is highly efficient.

The principle of the experiment is sketched on Fig. 1(a). More details can be found elsewhere [21]. Rubidium atoms effusing from oven $O$ are velocity-selected and promoted, one at a time, in the circular state $e$ (principal quantum number 51) in box $B$. The atom in the circular state crosses the superconducting cavity $C$, which sustains two non-degenerate orthogonally polarized modes $M_a$ and $M_b$. They have the same gaussian geometry with a $w = 6$ mm waist. Their damping times are 1.2 ms for $M_a$ and 0.9 ms for $M_b$. The $M_a - M_b$ frequency difference is $\delta/2\pi = 128$ kHz ($M_a$ has the highest frequency). At thermal equilibrium, $M_a$ and $M_b$ contain a thermal field with one photon on average. A field “erasure” procedure can be applied to prepare both modes in the vacuum state $|0\rangle$ [21]. An external source $S$ can be used to inject in $M_b$ a small coherent field with a tunable photon number. The modes are nearly resonant with the transition between circular states $e$ and $g$ (principal quantum number 50) at 51.1 GHz. The detuning $\Delta$ between the atomic frequency and $M_a$ can be tuned by changing the cavity length (slow tuning) or by Stark-shifting the atomic line (fast tuning). The final state of the atom is analyzed by the state-selective field-ionization detector $D$.

An atom entering an empty cavity in state $e$, resonant with $M_a$ ($\Delta = 0$) or $M_b$ ($\Delta = -\delta$), would undergo a quantum Rabi oscillation at the frequency $\Omega/2\pi = 49$ kHz [22]. We consider here a situation where the atomic transition is tuned above the frequency of mode $M_a$ ($\Delta > 0$). Direct photon emission in the cavity mode is thus forbidden by energy conservation, provided $\Delta$ is much larger than $\Omega$. 

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A higher order emission process in the initially empty target mode $M_a$ is however possible when the source mode $M_b$ initially contains a small field with a photon number distribution $p(n)$. Let us first consider the contribution of the $n$-photon Fock state in $M_b$. The initial system’s state, taken as the energy reference, is $|e, 0, n\rangle$. The symbols in the ket refer to the atomic state and to the $M_a$ and $M_b$ photon numbers successively. The initial state is coupled by photon emission in $M_a$ to $|g, 1, n\rangle$ (energy $-\hbar\Delta$) [see level scheme on Fig. 1(b)]. This level is coupled by a one-photon absorption in $M_b$ to $|e, 1, n - 1\rangle$ (energy $\hbar\delta$), coupled in turn to the final state $|g, 2, n - 1\rangle$ (energy $\hbar(\delta - \Delta)$). When $\Delta = \delta$, this third-order Raman process is resonant and injects two photons in $M_a$ using a single atom. Note that a symmetrical Raman process emits two photons in $M_b$ when $M_a$ contains a small field and $\Delta = -2\delta$. These Raman processes are related to the cascade in the dressed atom levels used in [23] to realize a two-photon optical laser oscillator with macroscopic atom samples. We are, however, interested here in a single step process involving only one atom.

The lowest order Raman coupling matrix element is $\Omega^3\sqrt{2n}/[8\Delta(\Delta + \delta)]$. The $\sqrt{n}$ and $\sqrt{2}$ factors arise from the stimulation of the $|g, 1, n\rangle \rightarrow |e, 1, n - 1\rangle$ and of the $|e, 1, n - 1\rangle \rightarrow |g, 2, n - 1\rangle$ couplings by the fields in $M_b$ and $M_a$ respectively. For a coherent or thermal source field, the two-photon emission probability is obtained by summing independently the various Fock state contributions in $M_b$, weighted by their probability $p(n)$. The efficiency of the process thus increases with the average photon number in the source mode.

This simple discussion neglects the light shifts produced by the non-resonant modes on the atomic levels. These second order effects are much larger than the Raman coupling. An effective hamiltonian model and a numerical simulation of the Raman emission process [24] show that these shifts reduce the $\Delta$ value at resonance, which becomes smaller than $\delta$ and decreases with the photon number in $M_b$ (for instance, for a six photon coherent field in $M_b$, the resonance condition is $\Delta/2\pi = 65$ kHz). For large source fields, the Raman transition merges with the ordinary emission line at $\Delta = 0$.

Figure 2 presents the probability $P_g$ for detecting the atom in state $g$, reconstructed by averaging 1000 realizations of the experiment, as a function of $\Delta$. The atomic velocity is
\( v = 200 \text{ m/s} \). The solid squares on figure 2(a) (with statistical error bars) correspond to both modes initially at thermal equilibrium. The main features are the two direct one-photon emission resonances at \( \Delta = 0 \) and \( \Delta = -\delta \). Two Raman resonances are also observed around \( \Delta/2\pi = 80 \text{ kHz} \) and \( \Delta/2\pi = -210 \text{ kHz} \). For the first, \( M_a \) is the target mode (see discussion above). For the second, \( M_a \) is the source and \( M_b \) the target mode. These features are reproduced by the numerical simulation of the evolution (solid line). The Raman process in this situation is produced by the thermal field in the source mode and enhanced by the initial thermal field in the target mode. These Raman resonances are not observed with fast atoms (above 300 m/s). The \( \Delta = 0 \) and \( \Delta = -\delta \) lines are wider in this case and the Raman transfer rate is much smaller. The constant 8\% background in figure 2 is mainly due to spontaneous emission outside the cavity, enhanced by the background thermal field.

The other data sets in figure 2(a) correspond to an initially empty \( M_a \), \( \Delta \) being scanned only around the upper Raman resonance. Open squares, obtained when \( M_b \) is empty, exhibit no resonant transfer. This demonstrates the essential role of the source field in the Raman process. The two other data sets correspond to a coherent field in \( M_b \) with an average photon number of 5.6 (open circles) and 11.2 (open squares). This field has been independently calibrated by a procedure described in [25]. These last two data sets are enlarged on figure 2(b), together with the results of the numerical simulations (dotted and solid lines). The simulations agree reasonably well with the experiment (note that the effect of the light shifts on the resonance frequency is here clearly apparent). For the largest source field, the transfer rate peaks at 30\%. For higher photon numbers, the Raman resonance merges into the one-photon line.

We have also directly probed the final field in \( M_b \). The same atom undergoes the Raman emission process and measures the field through the dispersive light shift experienced by the atomic level \( g \) [26]. The atom is initially prepared in \( e \), with a velocity \( v = 170 \text{ m/s} \). Mode \( M_a \) is empty and \( M_b \) contains a coherent field with 6 photons on the average. The cavity is initially tuned to the Raman resonance condition \( (\Delta/2\pi = 65 \text{ kHz}) \). The two-photon emission builds up. The frequency detuning \( \Delta/2\pi \) is suddenly increased from 65 kHz to 135
kHz, at a time $t = 5 \mu s$ after the atom has crossed the cavity axis. The Raman process is frozen from then on. The atom then undergoes a classical $\pi/2$ pulse which prepares a superposition of $g$ and $i$ (circular state with principal quantum number 49). This pulse, resonant on the $g \rightarrow i$ transition at 54.3 GHz, is fed from a classical source $S'$ into a low-Q transverse mode of the cavity structure. The phase of the atomic superposition is sensitive to the light shifts produced on $g$ by $M_a$ and $M_b$ (the $g \rightarrow i$ transition being far off-resonant with $M_a$ and $M_b$, the light shifts of level $i$ is negligible). These light shifts are proportional to the photon number in each mode and inversely proportional to the final values of $\Delta$ and $\Delta + \delta$. The final atomic superposition is probed by applying on the atom, after it exits $C$, a second $\pi/2$ pulse on the $g \rightarrow i$ transition, resulting in a Ramsey fringe pattern [21]. The probability for detecting the atom in $g$ is sinusoidal versus the frequency of $S'$. The phase of this modulation reflects the accumulated light shift [26]. Note that the final detection of the atom in $g$ or $i$ selects the events in which the Raman emission has taken place. If the atom is in $e$ at time $t$ (no Raman emission has occurred), it is unaffected by the Ramsey pulses. These events are discarded.

The corresponding fringes are plotted on figure 3 (solid squares), compared to two reference signals. The open square signal is obtained when $M_a$ is empty ($\Delta$ is detuned from the Raman resonance at all times). The phase of these fringes is defined as being zero. The open diamond signal corresponds to a one-photon Fock state in $M_a$. The Raman emission is replaced in this case by a resonant $\pi$ Rabi pulse in $M_a$. Note that, for the three data sets, $M_b$ initially contains the six photon coherent field. The phase of the fringes obtained after the Raman process and after the single photon emission are $\phi_2 = 0.52 \pm 0.04$ rd and $\phi_1 = 0.32 \pm 0.03$ rd respectively. The zero phase corresponds to the light shifts produced by the vacuum in $M_a$ and the 6 photon coherent field in $M_b$. The phase $\phi_1$ reflects the apparition of one photon in $M_a$ while $M_b$ remains unchanged. The phase $\phi_2$ results from the simultaneous change of fields in $M_a$ and $M_b$. The first field increases by two photons, thus producing a shift $2\phi_1$, and the second decreases by about one, thus reducing the phase by about $\phi_1/2$, since $\Delta + \delta \simeq 2\Delta$. With the precise detuning values, we predict $\phi_2/\phi_1 = 1.52$, \[ \phi_2 = 0.52 \pm 0.04 \text{ rd, } \phi_1 = 0.32 \pm 0.03 \text{ rd, } \phi_2/\phi_1 = 1.52, \]
in good agreement with the measured ratio $1.62 \pm 0.05$.

For a more quantitative analysis, we perform a numerical simulation of the experiment taking into account all known imperfections of the set-up: cavity relaxation, spontaneous emission events before or after the interaction with $C$, finite contrast of the Ramsey interferometer, two-atom events (the atom number has a Poisson distribution with a low average value [21]). The lines in figure 3 result from this simulation. They agree well with the experimental data. This confirms our interpretation of the emission process.

The results of this simulation can be used to determine the photon number distribution after the Raman emission process, at time $t$. The raw probability for having two photons is 37%. The broadening of the distribution is due to two main experimental imperfections. The $e \rightarrow g$ spontaneous emission before the atom enters the cavity generates a 20% background of events where the atom enters $C$ in $g$. This imperfection could easily be removed by emptying level $g$ immediately before the atom enters $C$ with a convenient microwave pulse, raising the two-photon probability up to 46%. The second imperfection is the growth of a one-photon residual thermal field in the cavity during the experiment. This background could be suppressed easily (the corresponding experimental improvements are in progress). The two-photon probability would raise then to 63%.

We have shown in this Letter that a single atom can prepare efficiently a two-photon Fock state in a cavity mode by a third-order Raman process. This opens the way to interesting extensions. A coherent superposition of zero- and two-photon Fock states can be readily obtained by preparing the atom initially in a superposition of $e$ and $g$. We have also observed, for $\Delta = \delta$, the reverse Raman process coupling $|g, 0, n\rangle$ to $|e, 1, n - 2\rangle$. The atom leaves a photon in $M_a$ and is simultaneously excited. This process removes two photon at the same time from the source mode $M_b$. A stream of atoms performing this Raman process could thus be used to implement an effective two-photon relaxation in mode $M_b$, opening interesting perspectives for relaxation tailoring and quantum states engineering.

With a continuous source of atoms, these Raman processes lead to a new type of micromaser [13]. We are now exploring theoretically its properties. It exhibits novel features,
compared to the one- and two-photon micromasers [13,24] or to the two-photon lasers [23]. Fock states with higher photon numbers can also be reached. When $\Delta = 2\delta$, a fifth order process resonantly couples $|e, 0, n\rangle$ to $|g, 3, n - 2\rangle$, injecting three photons in a single step into the target mode. With the atomic velocities available now, the corresponding transfer rate is too low (below $10^{-3}$) to be observed. The use of laser cooling techniques to prepare very slow circular atoms could allow the observation of this effect.

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FIG. 1. (a) Scheme of the experiment. (b) Energy diagram for the two-photon Raman emission.
FIG. 2. Probability $P_g$ for detecting the atom in $|g\rangle$ as a function of $\Delta$. (a) Solid squares: both cavity modes contain initially a thermal field. Error bars reflect the binomial detection statistics variance. Spontaneous emission resonances are observed at $\Delta = 0$ and $\Delta = -\delta$. The two Raman resonances are around $\Delta/2\pi = 80$ kHz and $\Delta/2\pi = -210$ kHz. The solid line results from a numerical simulation. Open squares: both modes initially empty. Open circles and diamonds: $M_a$ initially empty and $M_b$ containing 5.6 and 11.2 photon coherent fields respectively. (b) Enlargement of the Raman resonances for 5.6 (open squares) and 11.2 (open diamonds) photons in $M_b$. Lines result from a numerical simulation.

FIG. 3. Ramsey interference fringes on the $g \rightarrow i$ transition for three different states of the field in $M_a$. Open squares: reference fringes for $M_a$ in the vacuum state. Open diamonds: $M_a$ contains a single photon Fock state produced by a $\pi$ resonant spontaneous emission pulse. Solid squares: $M_a$ state results from a Raman transition. The lines are provided by a numerical simulation of the experiment (see text).