Strongly Interacting Photons in a Nonlinear Cavity

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We consider the dynamics of single photons in a nonlinear optical cavity. When the Kerr nonlinearities of *atomic dark resonances* are utilized, the cavity mode is well described by a spin-$1/2$ Hamiltonian. We show that it is possible to achieve coherent control of the cavity-mode wave function using π pulses for single photons that switch the state of the cavity with very high accuracy. The underlying physics is best understood as the nonlinearity induced anticorrelation between single-photon injection/emission events, which we refer to as *photon blockade*. We also propose a method which uses these strong dispersive interactions to realize a single-photon turnstile device. [S0031-9007(97)03903-3]

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It is well known that the observation of strictly quantum effects in optics relies on the existence of strong nonlinear interactions between photons. Examples of such quantum optical phenomena that have been demonstrated experimentally include quadrature squeezing by parametic down-conversion and measurement of nonlocal correlations of entangled photon states [1]. On the other hand, observation of quite a few fascinating phenomena such as the generation of a superposition of macroscopically distinct quantum states, i.e., *Schrödinger cats*, have been in most cases hindered by the difficulty in obtaining large nonlinearities in absorption (or decoherence) free media. To overcome this limitation, several groups have been studying nonlinear optics in the *strong coupling regime* where the coupled-system eigenstates are superpositions of (bare) atom-cavity states [2–4]. Recently, these Jaynes-Cummings nonlinearities have been utilized in the realization of a Schrödinger cat state in a microwave cavity [4].

Using an alternative approach, two of us (H.S. and A.I.) have recently proposed a new scheme which gives resonantly enhanced Kerr nonlinearities that are limited only by two-photon absorption [5]. These (essentially) absorption-free giant optical nonlinearities are obtained in the weak atom-photon coupling limit and can be extended to other material systems. Such a scheme can increase the available nonlinear phase shift by almost 10 orders of magnitude for a given light intensity and loss coefficient-length product of the atomic medium [5].

In this Letter, we consider the problem of a nonlinear optical cavity mode in the light of this new development and predict novel properties for both the intracavity and the transmitted light fields. The photon-photon interaction coefficient in such a nonlinear cavity could easily be much larger than the cavity decay rate and the linewidth of the driving field, implying that the cavity photons behave as strongly interacting particles. We show that the cavity-mode dynamics in this limit is well described by a spin-$1/2$ Hamiltonian. Using the analogy with atomic systems, we predict that it is possible to realize π pulses for photons which may be used to switch the state of the cavity with arbitrary accuracy. To explain the strong antibunching of transmitted photons, we introduce the concept of *photon blockade* in close analogy with the phenomenon of Coulomb blockade for quantum-well electrons. We also discuss a method which utilizes the strong dispersive interactions between photons to realize a single-photon turnstile device [6].

Figure 1 details the nonlinear cavity structure that we envision: A confocal cavity with finesse $\mathcal{F} \simeq 10^4$ contains a low density atomic medium, whose energy level structure may be represented by the four-state diagram shown in Fig. 2. A nonperturbative *coupling field* resonant with the $|2\rangle$-$|3\rangle$ transition creates an electromagnetically induced transparency (EIT) or a dark resonance [7] at the nth cavity-mode frequency, which is in turn assumed to be resonant with the $|1\rangle$-$|3\rangle$ transition ($\omega_{31} = \omega_{\rm cav}$). As a result, the cavity mode sees vanishing one-photon atomic loss but a giant self-phase modulation coefficient, provided that the atomic transition energies satisfy $\omega_{42} \simeq \omega_{31} \gg \omega_{21}$. The corresponding real part of the third-order nonlinear susceptibility is [5]

$$\mathrm{Re}[\chi^{(3)}] = \frac{N|\mu_{13}|^2|\mu_{24}|^2}{2\epsilon\hbar^3\Omega_c^2\Delta\omega_{42}}, \tag{1}$$

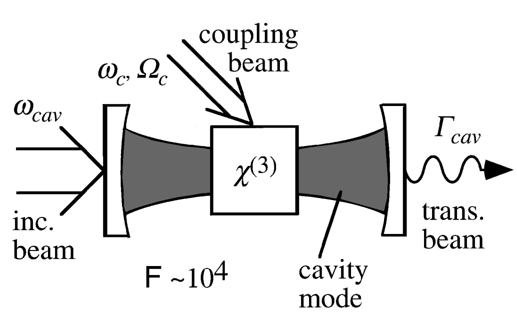

FIG. 1. The nonlinear cavity that we analyze. For simplicity, it is assumed that one of the mirrors has a smaller reflectivity and provides the output.

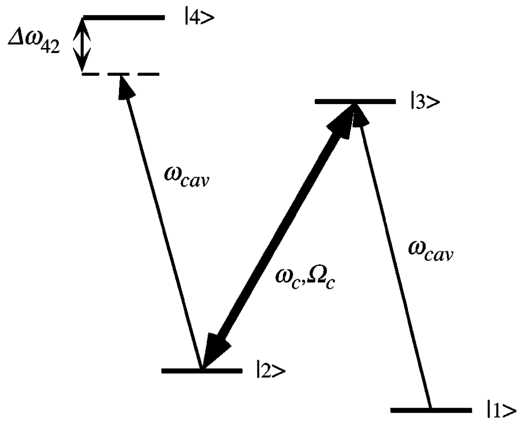

FIG. 2. The four-level atomic system that generates a resonantly enhanced two-photon absorption limited Kerr nonlinearity. We assume that the optical transitions $|3\rangle$-$|2\rangle$ and $|3\rangle$-$|1\rangle$ are resonant.

where N denotes the atomic density and μ_{ij} denotes the dipole matrix element of the $|i\rangle$-$|j\rangle$ transition. Ω_c is the Rabi frequency of the coupling field, and $\Delta\omega_{42} = \omega_{42} - \omega_{\rm cav}$. The principal feature of Eq. (1) is that the *effective detuning* from the first transition is given by Ω_c, which can be much smaller than the natural width of state $|3\rangle$. One can therefore obtain a large resonant enhancement in nonlinearity without creating any (single-photon) loss, provided that dephasing of the (hyperfine split) $|1\rangle$-$|2\rangle$ transition is negligible [5]. In steady state, all the atoms will be in the ground state $|1\rangle$ (with small virtual populations in the excited states $|2\rangle$ and $|4\rangle$, and none in state $|3\rangle$), implying that the system is two-photon absorption limited.

The dynamics of the cavity mode is best analyzed by adiabatically eliminating the atomic degrees of freedom. In the limit where the two-photon atomic losses are much smaller than the cavity decay rate, the effective Hamiltonian for the cavity mode ($\hat{a}$) driven by a classical light source is [1]

$$\hat{H}_{\rm eff} = \hbar\omega_{\rm cav}\hat{a}^\dagger\hat{a} + \hbar\kappa\hat{a}^\dagger\hat{a}^\dagger\hat{a}\hat{a} + i\hbar\sqrt{2\Gamma_{\rm cav}}\,(\beta^*\hat{a} - \beta\hat{a}^\dagger) - \frac{i\hbar\Gamma_{\rm cav}}{2}\,\hat{a}^\dagger\hat{a}\,, \quad (2)$$

where $\Gamma_{\rm cav} = \Delta\omega_{\rm ax}/\mathcal{F}$ denotes the cavity decay rate via mirror losses. The axial-mode spacing $\Delta\omega_{\rm ax}$ is assumed to be large compared to atomic and laser linewidths so that only one cavity mode needs to be considered. The third term describes the classical drive, where β is the electric field amplitude in natural units [1]. We assume that the laser amplitude is, in general, time dependent with a corresponding linewidth $\Delta\omega_{\rm laser}$. The reflectivity of the mirror on which the coherent field is incident is assumed to be higher than the other; however, we will still use a single decay/coupling coefficient $\Gamma_{\rm cav}$ and assume that the difference in reflectivities is incorporated into the *effective* amplitude β. The complete cavity dynamics is obtained after quantum fluctuations associated with the cavity decay are incorporated using stochastic wave-function methods [8]. The photon-photon interaction term κ can be obtained from $\mathrm{Re}[\chi^{(3)}]$,

$$\kappa = \frac{3\hbar\omega_{\rm cav}^2}{2\epsilon V_{\rm cav}}\,\mathrm{Re}[\chi^{(3)}] = 3\,\frac{|g_{13}|^2|g_{24}|^2}{\Omega_c^2\Delta\omega_{42}}\,n_{\rm atom}\,, \quad (3)$$

where $n_{\rm atom} = NV_{\rm cav}$ and $g_{ij} = (\frac{\omega_{ij}}{2\epsilon\hbar V_{\rm cav}})^{1/2}\mu_{ij}$.

We reiterate that the nonlinear cavity model described by the Hamiltonian of Eq. (2) was previously analyzed by several authors, and it was predicted that such a cavity would yield photon antibunching [9]. The novelty in our case is in the expression for κ which predicts photon-photon interaction coefficients that are orders of magnitude larger than what was previously considered possible (because of the one-photon loss limitation). Before presenting simulation results, we will make a rough numerical estimate of κ: We assume a 2 cm cavity with $V_{\rm cav} \simeq 1 \times 10^{-4}$ cm^3; a 1 cm atomic medium of density 3×10^{11} cm^{-3}; and typical dipole matrix elements for alkali atoms $\mu_{ij} \simeq 3 \times 10^{-29}$ C m. If we choose $\Delta\omega_{42} \simeq 2\Omega_c \simeq 1 \times 10^9$ rad/sec, we obtain a nonlinear interaction coefficient $\kappa \simeq 1 \times 10^8$ rad/sec. This is about 20 times larger than the cavity decay rate, provided $\mathcal{F} = 10^4$. For a typical upper state decay rate, $\Gamma_4 = 3 \times 10^7$ s^{-1}, the two-photon absorption rate is approximately 1/5 of the cavity decay rate, justifying the effective Hamiltonian of Eq. (2). Doppler broadening does not affect EIT (i.e., $\chi^{(1)}$) if states $|1\rangle$ and $|2\rangle$ are hyperfine split states [7]. Kerr nonlinearity is only slightly modified since typical Doppler widths are comparable to the assumed $\Delta\omega_{42}$. The principal effect of Doppler broadening in an atomic vapor cell is the enhancement of two-photon absorption ($\mathrm{Im}[\chi^{(3)}]$). Therefore, the assumption of negligible atomic losses is only valid for trapped atoms or beams. We note that Roch *et al.* [10] have recently demonstrated a magneto-optical trap with atomic density and volume that are comparable to those assumed here. Since the only nonperturbative coupling in our scheme is between two unoccupied atomic states, the trapping process remains unaffected.

The single-mode description of the cavity that we use is valid provided that the nonlinear interaction coefficient κ is much smaller than the axial-mode spacing. When $\kappa = 0$, the applied field couples all states in the harmonic ladder of cavity-mode eigenstates. On the other hand, for $\kappa \gg \Gamma_{\rm cav}, \beta\sqrt{\Gamma_{\rm cav}}, \Delta\omega_{\rm laser}$, the applied field may only couple the vacuum state $|0\rangle$ to the Fock state with a single photon $|1\rangle$ resonantly. The higher lying photon-number states may be neglected since they are out of resonance. The cavity mode in this limit is well described by the spin-1/2 Hamiltonian

$$\hat{H}_{\rm app} \simeq \hbar\omega_{\rm cav}\hat{\sigma}_{11} + i\hbar\sqrt{2\Gamma_{\rm cav}}\,(\beta^*\hat{\sigma}_{01} - \beta\hat{\sigma}_{10}) - \frac{i\hbar\Gamma_{\rm cav}}{2}\,\hat{\sigma}_{11}\,, \quad (4)$$

where we have introduced the projection operators $\hat{\sigma}_{ij} = |i\rangle\langle j|$ for photon-number eigenstates. Clearly, this effective model is analogous to an atom driven by a resonant laser field, where a two-level description is satisfactory despite the presence of infinitely many excited states. This is one of the principal results of our Letter: Using the nonlinearities of *atomic dark resonances*, we can create strongly interacting photons in a cavity mode. The dynamics of this cavity mode is in turn governed by the simple coherently driven spin-1/2 Hamiltonian of Eq. (4); this is justified by the numerical simulations we discuss below.

Before proceeding, we note that the *two-level behavior* of a strongly coupled atom-field system has been predicted in Ref. [2]: Tian and Carmichael have demonstrated that, when a classical drive field is tuned on resonance with one of the *vacuum Rabi resonances*, the atom cavity-mode *molecule* [2] oscillates coherently between the states $|g,0\rangle$ and $|l,1\rangle = (|e,0\rangle + |g,1\rangle)/\sqrt{2}$, where $|g\rangle$ ($|e\rangle$) denotes the atomic ground (excited) state and $|i\rangle$ denotes the Fock state with i photons. The principal difference between the spin-1/2 behavior that we predict and this previous result is that our predictions are obtained for a pure cavity mode by adiabatically eliminating the atomic degrees of freedom. This feature, together with the large number of atoms ($\simeq 1 \times 10^7$) participating in the nonlinear interaction, makes our scheme independent of atom-number fluctuations and decoherence in the atomic system. In addition, the realization of two-level behavior of a pure (nonentangled) cavity mode enables us to coherently control the cavity dynamics as we shall describe shortly.

The physics behind the effective Hamiltonian of Eq. (4) is best understood by considering the strong photon-photon interactions. When the cavity is in $|0\rangle$, a photon from the driving field is injected with a probability determined by the drive strength. However, injection of a second photon will be blocked, since the presence of two photons in the cavity will require an additional $\hbar\kappa$ energy, which cannot be provided by the incoming laser photons. Only after the first photon leaves the cavity can a second one be injected. The strong interactions between the photons therefore cause a *photon (Kerr) blockade* of cavity transmission, in direct analogy with the Coulomb blockade of resonant tunneling in mesoscopic semiconductors [11]. We remark that this discussion is valid for both thermal and coherent driving fields.

First, we consider the limit of a weak continuous-wave (cw) coherent driving field: Figure 3(a) shows the results of a single quantum Monte Carlo wave function (MCWF) simulation [8] for the atom-cavity parameters given earlier. We observe that the number of cavity photons varies between 0 and 1, but never exceeds unity due to the photon blockade effect. At times immediately following a photon loss event (via the imperfect cavity mirrors), the cavity mode is necessarily in the vacuum state. Therefore the detector that counts the photons emitted from the cavity will never register two photons in time intervals much shorter than the cavity decay time. The physics is completely analogous to that of a single (two-level) atom, where antibunching in resonance fluorescence has been observed. Figure 3(b) shows the normalized second-order coherence function $g^{(2)}(\tau)$, calculated using the MCWF method and the effective Hamiltonian of Eq. (2): We clearly observe that $g^{(2)}(0) = 0$, demonstrating the antibunching of the emitted photons. The dashed curve shows the same calculation carried out using the approximate Hamiltonian of Eq. (4): We observe in all cases that the predictions of the Hamiltonians are identical. The slight difference in $g^{(2)}(\tau)$ predicted by the two models is completely due to finite averaging effects; in fact, when we use the same set of random numbers for the two simulations, the corresponding curves are indistinguishable.

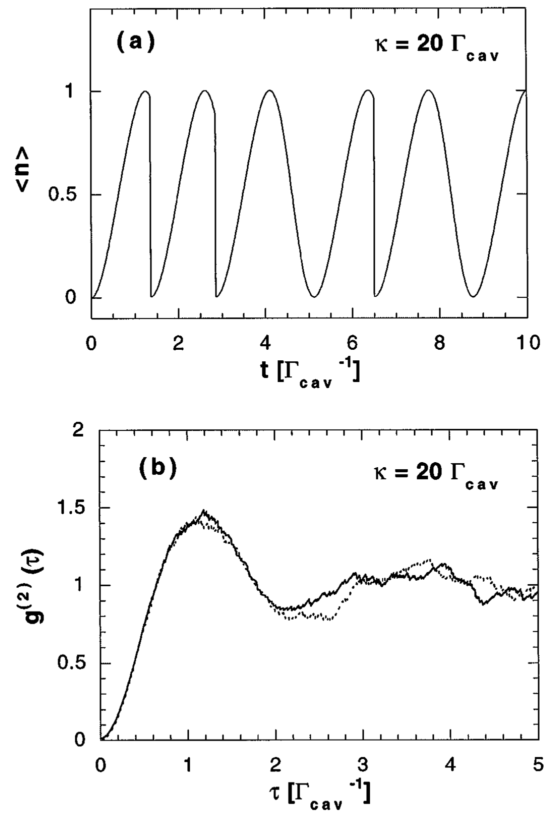

FIG. 3. MCWF simulation results under cw excitation: (a) the time evolution of the expectation value of the cavity-mode photon number for a single quantum trajectory, and (b) the second-order coherence function (solid line). The dashed line in (b) shows the same calculation carried out using the approximate spin-1/2 Hamiltonian.

Next, we consider the pulsed excitation of the cavity. It is well known in atomic physics that the state of a two-level atom can be set with arbitrary accuracy using coherent laser pulses that have a given $area = \int d\tau\,\Omega(\tau)$, where $\Omega(\tau) = 2\sqrt{2\Gamma_{\mathrm{cav}}}\,\beta$ is the time-dependent Rabi frequency. Since the nonlinear cavity that we are envisioning is governed by the same effective Hamiltonian as a

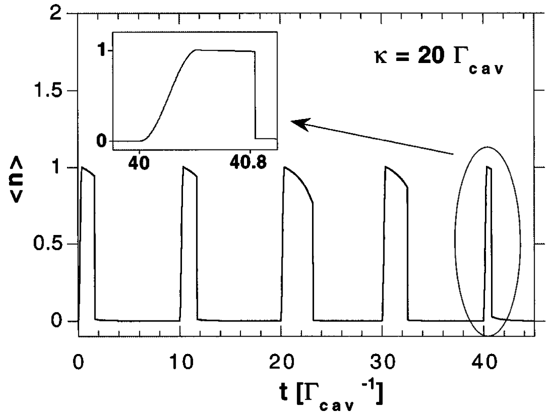

FIG. 4. MCWF simulation of the time evolution of the expectation value of the cavity-mode photon number under pulsed excitation. The plot shows a single quantum trajectory. For the chosen "area" of the classical drive field, one has π-pulse excitation of the cavity mode. (Inset: closeup of a single photonic π pulse.)

driven two-level atom, we expect to be able to switch the state of the cavity mode from $|0\rangle$ to $|1\rangle$ using a laser pulse of (dimensionless) area π. In fact, by adjusting this area, any coherent superposition $a|0\rangle + b|1\rangle$ of the two Fock states may be generated.

Figure 4 shows the time evolution of the cavity-mode photon number under pulsed laser excitation. Once again, simulations are carried out using the MCWF method with the effective Hamiltonian of Eq. (2). For simplicity, we have chosen square pulses with pulse width $\tau_{\text{laser}} = 0.4\Gamma_{\text{cav}}^{-1}$ and peak amplitude $\beta_{\max} = 2.8\sqrt{\Gamma_{\text{cav}}}$. We once again take $\kappa = 20\Gamma_{\text{cav}}$, which satisfies $\kappa \gg \tau_{\text{laser}}^{-1}, \sqrt{\Gamma_{\text{cav}}}\,\beta_{\max} > \Gamma_{\text{cav}}$. The quantum trajectory [8] result shown in the *inset* demonstrates that the cavity mode evolves from $|0\rangle$ to $|1\rangle$ after each applied laser pulse. By increasing κ and/or choosing optimal pulse shapes, one can further increase the degree of coherent control of the cavity wave function.

It is also shown in Fig. 4 that when a train of M such π pulses with a period $T_p = 10\Gamma_{\text{cav}}^{-1}$ is utilized, the generated output light field consists of optical pulses that contain one, and only one, photon. These heralded single photons in a given observation-time window form a special class of *multimode number states*: They have well-defined number and emission time information, which is achieved at the expense of increased phase and energy uncertainty. This is a nonclassical state of light with a characteristic second-order coherence function which, in the ideal case, exhibits peaks at $\tau = nT_p, n = 1, 2, \ldots$, and vanishes elsewhere [6]. The turnstile device described above has an uncertainty in the photon detection time that is given by Γ_{cav}^{-1}. An alternative approach, which can decrease the time uncertainty, is to introduce cavity dumping events that follow the π pulses by means of an intracavity modulator which decreases the reflectivity of one of the mirrors from ≈ 1 to ≈ 0. The generated heralded single photons in this case have a pulse width that is determined by the cavity round-trip time.

In summary, we have shown that resonantly enhanced absorption-free Kerrr nonlinearities open up a new domain for nonlinear optics in which the interaction strength of single photons is larger than all the other relevant energy scales. We have demonstrated that this property allows for the treatment of the cavity mode as a spin-$1/2$ system and opens up the possibility of realizing strong antibunching and deterministic photon injection into a cavity mode. The realization of a single-photon turnstile device and the generation of an arbitrary superposition of zero- and one-photon states may be useful in quantum computation applications.

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