Quantum analysis of the photonic blockade mechanism

Klaus M. Gheri, Wieland Alge, and Philippe Grangier*
Institut für Theoretische Physik, Universität Innsbruck, Technikerstrasse 25, A-6020 Innsbruck, Austria
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A recently proposed photonic blockade mechanism using four-level atoms inside an optical cavity is reviewed and analyzed. Various possible theoretical approaches are discussed, including adiabatic elimination of atomic variables, linearization, and numerical calculations in the weak excitation regime.
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Introduction. Driving multilevel atoms with several coherent light beams is a way to generate interesting nonlinear optical effects, either in the classical [1] or in the nonclassical regime of quantum optics [2]. Electromagnetically induced transparency (EIT), for instance, allows atoms irradiated by a laser beam to become transparent to a second light field on an otherwise optically thick transition [3] and can be used to obtain enhanced laser gain [4]. Interesting quantum-statistical properties have been predicted for light generated by devices based on EIT, such as narrower laser linewidths [5] and reduction of intensity fluctuations [6–8]. Photon turnstile devices have been recently realized in semiconductors [9]. In a recent Letter [10,11] Imamoğlu *et al.* have proposed an EIT-based scheme to create a giant Kerr nonlinearity with negligible absorption. Their scheme assumes that a large number N of atoms is *weakly* coupled to a single mode of an optical resonator. The authors have claimed that the collective interaction between the atoms and the resonator mode implements a photonic blockade mechanism: due to a large $\chi^{(3)}$ nonlinearity, a single photon inside the cavity creates a detuning large enough to prevent other photons from entering this cavity. This would implement a turnstile device for single photons, which could find application in the design of quantum logical gates for photonic quantum bits (qubits) [12] or in controlling the quantum noise of very-low-intensity light beams [13].

However, several of the assumptions made by Imamoglu *et al.* have been contested, including an adiabatic elimination procedure by which all atomic degrees of freedom [14,15] are removed. A preliminary semiclassical analysis [14] of the interaction, which does not employ adiabatic elimination, suggests that a dramatic reduction of available bandwidth for the light leaving the resonator should occur. Independently, Lukin *et al.* [16] have recently found that EIT within an optical cavity can give rise to a significant narrowing of the linewidth of the cavity light field. In this paper we will show that, beside a linewidth narrowing, the scheme also exhibits a critical dependence on the amount of energy that is temporarily stored in the atomic medium.

Model. The scheme presented in Ref. [10] relies on the interaction of an ensemble of N four-level atoms with a strong coherent driving field and a weakly coupled quantized single mode of the electromagnetic field inside a cavity (see Fig. 1, inset). The driving field is characterized by its Rabi frequency Ω and its optical frequency ω_p, taken equal to the transition frequency between levels $|1\rangle$ and $|2\rangle$. The action of the cavity field on the atoms is determined by the coupling constants g_1 and g_2 to the atomic transitions from levels $|0\rangle$ to $|2\rangle$ and $|1\rangle$ to $|3\rangle$, respectively. For simplicity we assume the empty cavity (resonance frequency ω_c) to be resonant with the transition between the atomic levels $|0\rangle$ and $|2\rangle$. The cavity mode is driven by a coherent field with amplitude $\alpha_{\rm in}/\sqrt{2\kappa}$ and center frequency $\omega_{\rm in}$, and losses through the single port of the cavity are accounted for by a damping rate 2κ. Spontaneous emission takes place from level $|2\rangle$ to levels $|0\rangle$ and $|1\rangle$ at the respective rates $2\Gamma_{02}$ and $2\Gamma_{12}$, and from level $|3\rangle$ to level $|1\rangle$ at a rate $2\Gamma_3$. The equations for the evolution of this system can be written conveniently in a rotating frame with respect to the external field frequencies $\omega_{\rm in}$ and ω_p. The Hamiltonian describing the *slow* system evolution is thus given by

$$\mathcal{H}(t)/\hbar = \sum_{\alpha=1,\ldots,N} [\mathcal{H}_s^\alpha + \mathcal{H}_{ra}^\alpha(t)] + \mathcal{H}_{rc}(t), \tag{1a}$$

$$\mathcal{H}_s^\alpha = \Delta_3\sigma_{33}^\alpha + \Delta_2\sigma_{22}^\alpha + \Delta_1\sigma_{11}^\alpha + (\Delta_c/N)a^\dagger a + \alpha_{\rm in}(a+a^\dagger)/N + (\Omega^*\sigma_{12}^\alpha + \text{H.c.}) + (g_1^* a^\dagger\sigma_{02}^\alpha + g_2^* a^\dagger\sigma_{13}^\alpha + \text{H.c.}), \tag{1b}$$

$$\mathcal{H}_{ra}^\alpha(t) = \sqrt{2\Gamma_3}b_3^\dagger(\alpha,t)\sigma_{13}^\alpha + \sqrt{2\Gamma_{02}}b_1^\dagger(\alpha,t)\sigma_{02}^\alpha + \sqrt{2\Gamma_{12}}b_2^\dagger(\alpha,t)\sigma_{12}^\alpha + \text{H.c.}, \tag{1c}$$

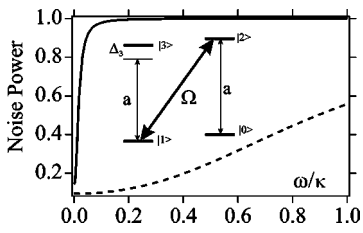

FIG. 1. Squeezing spectrum for the full (solid) and naive adiabatic (dashed) models demonstrating significant bandwidth narrowing. The inset shows the energy-level diagram of the system. Parameters: $\Omega=\Delta_3=30$, $g_1\alpha_s=0.1$, $\Gamma_{j2}=\Gamma_3=1$, $\kappa=0.1$, and $\mu=111$.

*Permanent address: Institut d'Optique Théorique et Appliquée, B.P. 147, 91403 Orsay Cedex, France.

$$\mathcal{H}_{rc}(t)=\sqrt{2\kappa}b^{\dagger}(t)a+\text{H.c.}, \tag{1d}$$

where the following detunings have been defined:

$$\Delta_c=\omega_c-\omega_{\text{in}}, \quad \Delta_3=\omega_3+\omega_p-2\omega_{\text{in}},$$

$$\Delta_2=\omega_2-\omega_{\text{in}}, \quad \Delta_1=\omega_1+\omega_p-\omega_{\text{in}},$$

with ω_j being the energy differences between states $|j\rangle$ and $|0\rangle$. In the remainder of this paper we will assume that Ω and the cavity field form a Raman resonance, i.e., $\Delta_1-\Delta_c=0$.

The reservoir operators obey usual bosonic commutation relations, and since the reservoirs are in a vacuum state, we are in the limit of *quantum white noise*, and the noise operators are δ correlated in time. This allows us to describe the evolution of the atom-cavity system operators by using the usual damping and fluctuation terms. We note that the Hamiltonian (1) couples all atoms with identical coupling strength to the cavity mode a and to the coherent driving field Ω, respectively. This symmetry of the dynamics would allow us to restrict the time evolution to the fully symmetric subspace of the atomic Hilbert space. However, fluctuations of the coupling strength (due to atomic motion) and spontaneous emission will prevent the atoms from acting in an exclusively cooperative fashion. In order to account for this, we will adopt the *independent-atom approximation*, which assumes that atom-atom correlations are negligible. Spontaneous emission is supposed to take place independently in each atom. This is known as the *private bath assumption* and is consistent with the approximation of uncorrelated atoms. It amounts to neglecting all collective spontaneous radiative events other than those mediated through the cavity mode. A brute force solution of the system's evolution remains very difficult, due to both the nonlinear character of the Heisenberg equations of motions and the huge dimension of the system's Hilbert space. We therefore have to adopt further simplifying assumptions as to the system's effective size or to the nature of its time evolution.

Solution strategies. Adiabatic elimination of the atomic degrees of freedom is the method chosen by Imamoglu and co-workers. Their basic assumption was that the atomic internal states evolve rapidly on the time scales relevant for the cavity mode. The mode evolution equation for the unapproximated model reads

$$da(t)=dLa(t)-i\sum_{\alpha=1,\ldots,N}[g_1^{*}\sigma_{02}^{\alpha}(t)+g_2^{*}\sigma_{13}^{\alpha}(t)]dt, \tag{2}$$

$$dLa(t)=-[(\kappa+i\Delta_c)a(t)+i\alpha_{\text{in}}]dt-i\sqrt{2\kappa}\,dB(t),$$

where $dB(t)=B(t+dt)-B(t)=b(t)dt$. Adiabatic elimination of the atoms allows us to express the coherences appearing in Eq. (2) in terms of the mode operators. Formally this is accomplished by neglecting the time derivatives of the atomic coherence operators σ_{ij}^{α}. To obtain a Taylor series expansion in powers of the mode creation and annihilation operators up to third order, one sets for $i\leqslant j$,

$$\sigma_{ij}^{\alpha}\approx p_{ij,\alpha}^{(0)}+p_{ij,\alpha}^{(1)}a+p_{ij,\alpha}^{(2)}a^{\dagger}a+p_{ij,\alpha}^{(3)}a^{\dagger}a^2. \tag{3}$$

Since the mode couples weakly to each individual atom, $p_{00,\alpha}=1$ is a meaningful assumption to iteratively determine the remaining coefficients of the expansion. Reinsertion of the appropriate terms into the evolution equation of the mode leaves us with a parametric model (Kerr effect) for the intracavity interaction with a nonlinear coupling strength proportional to the number of atoms. The approximated mode equation then reads

$$da=dLa(t)+i\chi^{(3)}a^{\dagger}a^2dt-2i\sqrt{|\text{Im}\,\chi^{(3)}|}a^{\dagger}dB_{\text{ex}}(t), \tag{4}$$

with $\text{Im}\,\chi^{(3)}>0$ and dB_{ex} representing vacuum excess noise due to two-photon absorption. This equation, however, disregards two essential aspects of the problem. Since EIT causes the otherwise dominant linear dispersive and absorptive contribution in the expansion of σ_{02}^{α} to disappear, we are left with a third-order nonlinearity as the leading term. This effect is, however, strongly frequency dependent, and any slightly mistuned frequency component of the mode will experience huge linear dispersion. Since the nonlinearity must at least be comparable to the cavity damping constant κ for an appreciable nonlinear effect, all frequency components that are not extremely close to the cavity resonance frequency will be driven out of resonance and are strongly suppressed. We thus conclude that the temporal response of the mode will be poorly described by Eq. (4). Another problematic aspect is that the field contains less than one photon on average, so that shelving of energy in the atomic medium must be expected to degrade the statistics. Again all such effects have been disregarded in the adiabatic elimination procedure adopted in Ref. [10].

In order to demonstrate the bandwidth narrowing [14], we assume that the mode contains a sufficiently large number of photons that a field with a reasonably well-defined coherent amplitude is established in the resonator. This allows us to test the validity of the adiabatic approximation, as the same physical mechanism is still at work, despite the different nature of the field statistics. Instead of a regular source of single photons this scheme offers the possibility of efficiently suppressing the quantum fluctuations in a phase quadrature component of the light leaving the resonator. The independent-atom assumption suggests the introduction of rescaled collective atomic operators: $\sigma_{ij}=\Sigma_{\alpha}\sigma_{ij}^{\alpha}/\text{N}$. These rescaled quantities, together with the field operators, now form a set of 18 operators whose time evolution equations are directly obtainable from Eq. (1). A quantum noise analysis of the system can be obtained by linearizing the 18 Itô quantum stochastic differential equations around their semiclassical mean values, i.e., by writing $X(t)\approx\overline{X}+\delta X(t)$ for any system operator X with δX being small [17–19,22,23]. We thus obtain two sets of equations. The first one is nonlinear and comprises only the semiclassical steady-state expectation values $\overline{X}$. For instance, the equation describing the macroscopic behavior of the cavity light field $\overline{a}=\alpha$ reads

$$\partial_t\alpha=-(\kappa+i\Delta_c)\alpha-iN(g_1^{*}\overline{\sigma}_{02}+g_2^{*}\overline{\sigma}_{13})-i\alpha_{\text{in}}. \tag{5}$$

The fluctuation terms $\delta X(t)$, where X can be any of the 18 system operators, form a linear set of differential equations that describe the atom-cavity response to small deviations

from the semiclassical steady state. The equation for the deviations from the steady-state expectation value α_s of a is given by

$$d\,\delta a = dL\,\delta a(t) - iN(g_1^*\,\delta\sigma_{02}(t) + g_2^*\,\delta\sigma_{13}(t))dt. \quad (6)$$

The way to deal with such linearized equations is standard and will not be detailed here. With three-level systems [24], the utility of such an analysis has been underpinned by the excellent agreement between theoretical prediction and experimental result in a recent quantum nondemolition experiment [13].

Discussion. In order to get dispersive effects with negligible absorption, one expects that the atoms should remain in the ground state $|0\rangle$ with a probability close to unity. To obtain the semiclassical steady-state solution, we thus follow the logic used in Eq. (3). Up to third-order perturbation theory in the field-coupling constants g_1 and g_2, the coherences are given by the following expressions ($\Delta_2 = 0$):

$$\bar{\sigma}_{01} \approx -\frac{g_1\alpha_s\Omega^*}{|\Omega|^2}, \quad \bar{\sigma}_{11} \approx \frac{|g_1\alpha_s|^2}{|\Omega|^2}, \quad (7a)$$

$$\bar{\sigma}_{03} \approx i\frac{g_1 g_2 \alpha_s^2 \Omega^*}{|\Omega|^2(\Gamma_3 + i\Delta_3)}, \quad (7b)$$

$$\begin{bmatrix}\bar{\sigma}_{02}\\ \bar{\sigma}_{13}\end{bmatrix} \approx -i\alpha_s\begin{bmatrix}g_1\\ g_2\end{bmatrix}\frac{|g_2\alpha_s|^2}{(\Gamma_3 + i\Delta_3)|\Omega|^2}. \quad (7c)$$

We realize that the average collective interaction term experienced by the mode amplitude is given by

$$-iN(g_1^*\bar{\sigma}_{02} + g_2^*\bar{\sigma}_{13}) = -\alpha_s|\alpha_s|^2 2\mu(\Gamma_{\rm ad} + i\Delta_{\rm ad}), \quad (8)$$

with $(\Gamma_{\rm ad}, \Delta_{\rm ad}) = (\Gamma_3, -\Delta_3)|g_2|^2/(\Gamma_3^2 + \Delta_3^2)$ and $\mu = |g_1|^2 N/|\Omega|^2$. In the limit of $\Gamma_3/|\Delta_3| \ll 1$, this term is predominantly dispersive in nature. Note that, as claimed by Imamoglu and co-workers, there is no contribution linear in the field amplitude due to EIT.

Further calculations are now best carried out in the frequency domain; we thus define Fourier transforms according to $\delta a(\omega) = \int dt\,\exp(i\omega t)\delta a(t)/\sqrt{2\pi}$. Then the Taylor series expansion outlined above also carries over to the fluctuations in the atomic coherences. Here we only wish to consider the two relevant atomic fluctuation terms $\delta\sigma_{02}(\omega)$ and $\delta\sigma_{13}(\omega)$. The notable difference from the previous case is that we already obtain a contribution to $\delta\sigma_{02}(\omega)$ that is linear in $\delta a(\omega)$:

$$-g_1^* N\,\delta\sigma_{02}(\omega) \approx \omega\,\delta a(\omega)\mu/[1 - (i\omega\Gamma_2 + \omega^2)/|\Omega|^2]. \quad (9)$$

with $\Gamma_2 = \Gamma_{02} + \Gamma_{12}$. Note that this term vanishes at zero-frequency. The zero-frequency limit reproduces exactly the result from an adiabatic elimination by simply neglecting any frequency dependence of the atomic coherences. This term can, however, become extremely large, as we may gather from its linear dependence on the number of atoms. For it to remain negligible, one requires $\mu = |g_1|^2 N/|\Omega|^2 \ll 1$. This was the main result obtained in Refs. [14,15]. The main effect of the linear dispersive term is to narrow the available frequency window. The narrowing becomes apparent from Fig. 1, where we plot a squeezing spectrum obtained for parameters close to the ones used in Ref. [10]. The noise reduction bandwidth is drastically smaller than the cavity bandwidth ($\mu = 111$).

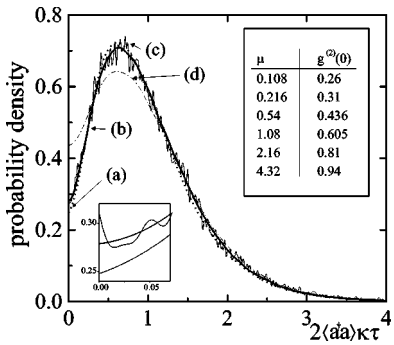

FIG. 2. Normalized waiting time distributions obtained from Eq. (11) for (a) $\Delta_{\rm ad}\mu = 2.16\kappa$, $\alpha_{\rm in} = 0.5\kappa$, $\mu = 0$ (this reproduces the naive adiabatic model with $|{\rm Re}\,\chi^{(3)}| = 2.16\kappa$), (b) $\mu = 0.108$, and (d) $\mu = 0.5$. Curve (c) is obtained from a 30-atom quantum–Monte Carlo simulation for parameters that are expected to yield the same effective parameters as those for curve (b). Clearly curve (b) with $\mu = 0.108$ provides a better fit than curve (a) with $\mu = 0$. The inset shows the dependence of $g^{(2)}(0)$ on μ as obtained from a full one-atom model (four internal levels) with $\Delta_{\rm ad}\mu = 2.16\kappa$ kept constant and μ being adjusted by varying g_1.

To account for energy storage effects and bandwidth reduction on the single-photon level, we adopt an alternative description based on dark dressed states. In a first step we describe the presence of N atoms by a single effective atom with rescaled coupling constant $g_1 \to g_1\sqrt{N}$. Preliminary results from genuine many-atom calculation indicate that such a description is meaningful in the parameter regime considered by Imamoglu *et al.* Next we make he following ansatz for the wave function of the atom-cavity system:

$$|\Psi\rangle = \left[\frac{g_1\sqrt{N}}{\Omega}a|1\rangle - |0\rangle\right]\left(\frac{\Lambda}{\mu}\right)^{-1/2}|\Psi'\rangle, \quad (10)$$

with $|0\rangle$ and $|1\rangle$ denoting the two ground states of the effective atom, and $\Lambda = \mu/(\mathbb{1} + \mu a^\dagger a)$. Equation (10) assumes that the coupled atom-cavity systems remains in a state that is commonly known as a ''noncoupled'' or dark state [21]. By allowing only such superposition states we automatically eliminate the exited atomic state $|2\rangle$ from our description. This implies that the auxiliary normalized wave function of the cavity field state $|\Psi'\rangle$ satisfies

$$\begin{aligned} d|\Psi'\rangle = [&-(\kappa + i\Delta_c)a^\dagger a(\mathbb{1} - \Lambda) - (\Gamma_{\rm ad} + i\Delta_{\rm ad})a^{\dagger 2}a^2\Lambda \\ &- i\alpha_{\rm in}(\Lambda^{-1/2}a\Lambda^{1/2} + \Lambda^{1/2}a^\dagger\Lambda^{-1/2})]|\Psi'\rangle dt \\ &- i(\sqrt{2\kappa}\Lambda^{-1/2}a\,dB^\dagger + \sqrt{2\Gamma_{\rm ad}}a^2\,dB_{\rm ex}^\dagger)\Lambda^{1/2}|\Psi'\rangle. \end{aligned} \quad (11)$$

Imamoglu's adiabatic model, cf. Eq. (4), can be recovered as the limit where $\mu \to 0$ and $(i\Gamma_{\rm ad} - \Delta_{\rm ad})\mu \to \chi^{(3)}/2$. This description is valid in the adiabatic limit; i.e., the internal atomic evolution is much faster than that of the mode (κ, $\alpha_{\rm in}$). We also assume that $\Delta_1 = 0$ and that $\Delta_{\rm ad}$

$\ll \max(\Omega, \sqrt{N} g_1)$. Our model now takes into account that a small fraction of the atoms can be pumped into their internal states $|1\rangle$. This is an important effect if the field contains only a small number of photons. The extent to which this takes place is given by the size of μ. We found a strong dependence of $g^{(2)}(\tau)$ [25] on μ, with already small values of $\mu \approx 0.5$ giving rise to a significant degradation of the achievable antibunching; cf. Fig. 2. Only for $\mu \ll 1$ is the effect slight. In this limit we have also been able to show that the time evolution of the average photon number is slowed down $[t \rightarrow t/(1+\mu)]$, as also predicted by the previously discussed semiclassical model (bandwidth narrowing). A comparison with a density-matrix solution for a full (all states taken into account) rescaled one-atom model yields excellent agreement (we have tested this for values of μ up to 2). Consequently, for the scheme to work the following minimum constraints have to be satisfied:

$$\mu \ll 1, \quad |\Delta_{\mathrm{ad}}| \mu / \kappa > 1. \tag{12}$$

The second condition implies $|\Delta_{\mathrm{ad}}|/\kappa \gg 1$ and thus $|g_2| \gg \kappa$. The optimal choice of $\Delta_3 = \Gamma_3$ still requires $|g_2| \gg \sqrt{2\Gamma_3 \kappa}$ [26,27]. This clearly undermines the attractiveness of the original proposal, whose appeal was confounded in the seeming absence of a need for strong coupling. Yet the present proposal could work in the limit of strong coupling with a mesoscopic number of atoms. Werner and Imamoglu have come to a similar conclusion in a recent manuscript [28] on the same system in an off-resonant configuration $(\Delta_1 - \Delta_c \neq 0)$. Our description assumes that the behavior of a single atom does not significantly change the properties of the cavity field. Since the field considered here comprises only a few photons, the interaction of all atoms with the mode may depend on whether or not a photon is lost to the environment in a spontaneous-emission event. This issue still requires further work [27].

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