Nonclassical Intensity Correlations in Cavity QED

S. L. Mielke, G. T. Foster, and L. A. Orozco

Department of Physics and Astronomy, State University of New York, Stony Brook, New York 11794-3800

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We report nonclassical correlations in the light transmitted by a collection of two-level atoms strongly coupled to a single mode of the electromagnetic field of a driven optical cavity. This cavity QED system can produce bunched light that violates the Schwarz inequality due to the state preparation of the system from the detection of a photon. The correlation function shows oscillations at a frequency that *decreases* with increasing intensity. [S0031-9007(98)05998-5]

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An ideal environment to study nonclassical electromagnetic fields is a system consisting of one or more two-level atoms coupled to a single mode of the electromagnetic field of an optical resonator. This system, originally studied by Jaynes and Cummings [1], has become the arena of cavity quantum electrodynamics (QED). Recent explorations of the effects of strong coupling between atom and cavity include the vacuum Rabi splitting for an atom in an optical cavity [2], the micromaser [3], the microlaser [4], and the temporal evolution of both a microwave field as probed by the state of a Rydberg atom [5] and the analog system of an ion in a rf trap [6]. The observation and measurements of the second order intensity correlation by Rempe *et al.* [7] showed the nonclassical effect of antibunching in cavity QED. In this Letter we describe measurements of the second order correlation function in cavity QED. The correlation function is a probe of the dynamics that relies explicitly on quantum mechanical ideas of interference and collapse of the wave function. We have found states of the electromagnetic field that violate the Schwarz inequality in a regime not previously explored.

The second order correlation function of the intensity $g^{(2)}(\tau) = \langle \hat{I}(t)\hat{I}(t+\tau)\rangle / \langle \hat{I}(t)\rangle^2$ (normal ordered and time ordered) is related to intensity fluctuations and their enhancement or suppression relative to an ideal laser field for which $g^{(2)}(\tau) = 1$. The correlation function contains information about the probability of detecting one photon at time $t + \tau$ given that one was detected at time t. From the Schwarz inequality there are two conditions on the correlation function for classical fields [8]: First $g^{(2)}(0) > g^{(2)}(\tau)$ and second $|g^{(2)}(0) - 1| > |g^{(2)}(\tau) - 1|$. Antibunching $[g^{(2)}(0) < g^{(2)}(0^+)]$ [9,10] and sub-Poissonian statistics $[g^{(2)}(0) < 1]$ are intimately related to the first condition. The second has not been explored as thoroughly. In this Letter we report a strong violation of the latter inequality in our system. The violation of either of them implies a nonclassical electromagnetic field. Quantum correlations are required to explain such features.

Two dimensionless numbers characterize cavity QED systems: the single atom cooperativity C_1 and the saturation photon number n_0. They scale the influence of an atom and the influence of a photon in the system. These two numbers relate the reversible dipole coupling of a single atom with the cavity mode (g_0) with the irreversible coupling to the reservoirs through cavity (κ) and atomic polarization and inversion decays ($\gamma_\perp, \gamma_\parallel$) by $C_1 = g_0^2/2\kappa\gamma_\perp$ and $n_0 = 2\gamma_\perp\gamma_\parallel/3g_0^2$. In the strong coupling regime of cavity QED, $C_1 > 1$ and $n_0 < 1$.

The spectroscopic structure of N effective atoms coupled to a cavity mode evolves from a harmonic doublet, or vacuum Rabi splitting, to a singlet when the intensity (number of photons n) is sufficiently high ($n > n_0$). This system has been extensively studied in the context of optical bistability (OB) [11] and cavity QED [12]. The field inside the cavity x, normalized by $\sqrt{n_0}$, is related to the normalized driving intensity Y by the relation from OB for low x, $Y \approx (1 + 2C_1N)^2|x|^2$ [11].

Previous measurements of $g^{(2)}(\tau)$ examined antibunching in the intensity region of the harmonic doublet [7]. Here we present nonclassical correlation functions for stronger coupling parameters. Two violations of the Schwarz inequality are reported: $g^{(2)}(\tau) > g^{(2)}(0)$ in the presence of both bunching and super-Poissonian statistics $[g^{(2)}(\tau) > g^{(2)}(0) > g^{(2)}(0^+) > 1]$, and antibunching with super-Poissonian statistics $[g^{(2)}(0^+) > g^{(2)}(0) > 1]$. We also report measurements of the intensity dependence of the oscillation frequency of $g^{(2)}(\tau)$.

Carmichael *et al.* [13] studied the collapse of the wave function in cavity QED. They pointed out the factorization of the correlation function, such that $g^{(2)}(\tau) = \langle \hat{I}(\tau)\rangle_c / \langle \hat{I}(t)\rangle$, where $\langle \hat{I}(\tau)\rangle_c$ is the transient response of the cavity intensity following the quantum jump made when the wave function collapses as a photon leaves the cavity at $\tau = 0$. For $\tau \geq 0$ and very low intensities, the result is

$$g^{(2)}(\tau) = |1 + \mathcal{A}\mathcal{F}(\tau)|^2. \quad (1)$$

$\mathcal{F}$ has a damped oscillation at the vacuum Rabi frequency: $\mathcal{F}(\tau) = \exp(-\beta\tau)[\cos(\Omega_0\tau) + (\beta/\Omega_0) \times \sin(\Omega_0\tau)]$ with $\Omega_0 = \sqrt{g_0^2 N - [(\kappa - \gamma_\perp)/2]^2}$ for N effective atoms [7] and a decay rate of $\beta = (\kappa + \gamma_\perp)/2$. $\mathcal{A}$ is the relative change of the field inside the cavity caused by the sudden escape of one photon [13]: $\mathcal{A} = -4C_1^2 N/[(1 + \gamma_\perp/\kappa)(1 + 2C_1N) - 2C_1]$. The jump occurs because the polarization of the medium increases when a photon leaves the cavity. The collective cavity enhancement of the dipole decay rate is reduced in the ratio

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$(N-1)/N$ and this increases the polarization amplitude (which is inversely proportional to the damping rate).

For strong coupling ($C_1 > 1$), the change can be many times larger than the intracavity field. All of the effects in this system are largely independent of N since, for large N, $\mathcal{A}$ is proportional to C_1: $\mathcal{A} \approx -2C_1/(1+\gamma_\perp/\kappa) + \mathcal{O}(1/N)$.

From this dependence on $\mathcal{A}$, $g^{(2)}(0)$ diverges as C_1 goes to infinity. Photons escape in pairs (or more generally in clumps) because the cavity is filled with a saturable absorber. When the coupling constant is large the saturation effect can be turned on by single photons. One photon in the cavity makes the medium transparent and more photons come through. This is the same mechanism that switches absorptive bistability to the upper branch. Here, however, the saturation occurs for very low mean photon numbers and the shot noise in the incident photon stream prevents a mean saturation from being sustained.

An important prediction of this model [13] is the existence of intensity correlation functions that oscillate at the vacuum Rabi frequency and its harmonics and have larger amplitudes for the oscillations after a peak at $\tau = 0$. These correlations strongly violate the second Schwarz condition and are purely quantum mechanical in nature. Physically, the detection of the first photon prepares a polarization in the system sufficiently large to change the sign of the cavity field [13]. This polarization then radiates into the cavity with characteristic delay time of half an oscillation. Our measurements show clear evidence of these dynamics.

Figure 1 shows a schematic of our apparatus. Two 0.78 cm diameter high reflectivity mirrors form a standing wave optical cavity with Gaussian transverse mode. We use different transmission coefficients ($2.3\times10^{-4}, 2.3\times10^{-5}$), radii of curvature (5, 17.5, and 20 cm), and mirror separations d (789, 325, and 175 μm) to obtain a range of g_0 and κ in the strong coupling regime. The maximal coupling between an atom and the mode is $g_0 = (\mu^2\omega/2\hbar\epsilon_0 V)^{1/2}$; μ is the transition-dipole moment of the atom, ω is the resonance frequency of both atoms and cavity, and $V = \pi w_0^2 d/4$ the cavity mode volume with waist w_0. An oven heated to 430 K produces an effusive beam of highly collimated Rb atoms ($\gamma_\parallel^{-1} = 26.2\times10^{-9}$ sec). The atoms are optically pumped before intersecting the cavity mode at 90°. A liquid nitrogen cooled surface surrounds the interaction region reducing any background atomic vapor. The excitation source is a cw titanium sapphire laser locked on resonance to the $5S_{1/2}, F=3 \rightarrow 5P_{3/2}, F=4$ transition of ^{85}Rb at 780 nm. The laser beam is split into an intense auxiliary beam to lock the cavity on resonance using FM sidebands and a weaker signal beam. A chopper wheel alternatively passes the lock (C1) or the signal (C2) beam at a rate of $\approx$1 kHz. We direct the output of the cavity into a 50/50 beam splitter and detect the photons with avalanche photodiodes (APD), EG&G SPCM-AQ-151. Detection of a photon by APD 1 starts a Lecroy 3377 time to digital converter that records the time of detection of up to sixteen consecutive photons by APD 2. There are interference filters and polarizers in front of each detector that suppress the problem of photons emitted by the APDs during the avalanche process [14]. We build up a histogram of photoelectric coincidences for different time delays from -400 ns to $+600$ ns from the start photon. Typical counting rates for the detectors are 30 kHz with a background count rate of less than 1 kHz. For a given input intensity we collect data for approximately 30 min. During this time the cavity and the laser remain locked with variations of less than $\pm\kappa/3$ and $\pm\gamma_\perp/3$. Veto electronics reject data collecting when the lock of the cavity is outside the range.

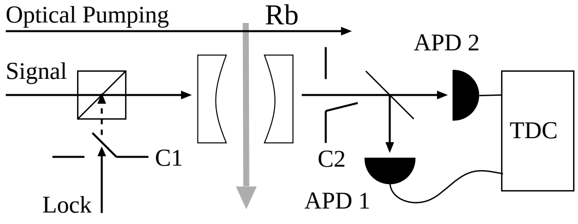

FIG. 1. Simplified diagram of the experimental setup.

We characterize the system by two measurements: First we measure the low intensity doublet of the vacuum Rabi splitting by transmission spectroscopy to obtain Ω_0. Second, we find the input intensity Y_{crit}, where the two sidebands merge when the intensity is high [15]. We identify this point by the joining of the two anharmonic peaks into a single broad feature. This is the entrance to the region of resonant intensity optical bistability.

When the system is in the strong coupling regime $C_1 = 7.4$ we see a correlation function that shows bunching followed by oscillations, with a large increase in the amplitude after half a cycle. Figure 2 shows an example of $g^{(2)}(\tau)$ for $C_1 = 7.4$, $n_0 = 0.1$, $Y/Y_{\mathrm{crit}} = 0.2$, and $N \approx 10$. We also show the sensitivity of this effect to atomic detunings between the laser frequency ω_l and the atomic transition ω_a. The detuning is given in normalized form $\Delta = (\omega_a - \omega_l)/\gamma_\perp$. The shaded region in Fig. 2(a) gives the allowed region for $g^{(2)}(\tau)$ from the second Schwarz condition: $|g^{(2)}(0) - 1| > |g^{(2)}(\tau) - 1|$. A $g^{(2)}(\tau) \approx 3$, well outside this range, is observed in Fig. 2(b) while a detuning of half a linewidth destroys the violation. This effect is related to the coherent emission of the collective dipole moment once one atom is no longer part of it. The detection of the first photon projects the system into a quantum state that then evolves showing a correlation function with distinctive nonclassical features. The observation of this novel effect is in qualitative agreement with the predictions by Carmichael *et al.* [13], despite the fact that we are not operating in the low intensity regime. At lower intensities we observe the correlation function oscillating around unity but the statistics are very poor.

The correlation measurements in Fig. 2 show that the detection of a first photon produces a high atomic

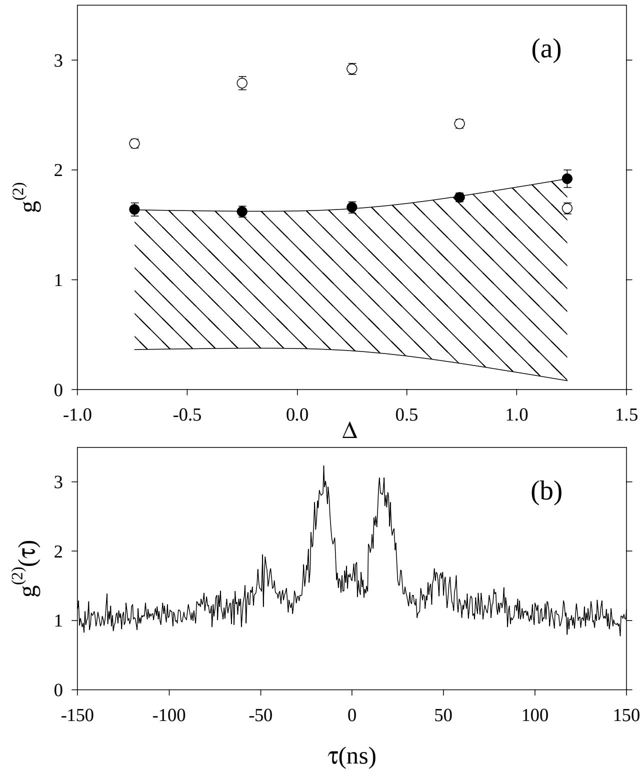

FIG. 2. (a) $g^{(2)}(\tau)$ at $\tau = 0$ (filled dots) and at $\tau \approx \Omega_0^{-1}/2$ (circles) as a function of atomic detuning Δ ($C_1 = 7.4$, $n_0 = 0.1$, $N \approx 10$, and $Y/Y_{\text{crit}} = 0.2$). The shaded region gives the values allowed by the Schwarz inequality. (b) $g^{(2)}(\tau)$ with $\Delta = 0.25$ showing bunching and the nonclassical behavior that violates the Schwarz inequality.

polarization. The system then evolves towards a steady state in a similar manner to its response to a step change in the driving field. We found the same intensity dependence in measurements of the oscillatory response of the system to step excitation [16], but the specific evolution is different in that case, since in the correlation measurement the step is in the polarization while in our previous measurements it is in the driving field. We do not have a theory for quantitative comparisons with Fig. 2 since existing calculations depend upon either large N expansions (weak coupling regime) or low intensity.

Figure 3 shows the evolution of the photon statistics for a system with $C_1 = 3.9$ as the intensity increases. $g^{(2)}(0)$ changes from <1 (sub-Poissonian) to >1 (super-Poissonian). All along the way, the field also shows antibunching, $[g^{(2)}(0) < g^{(2)}(0^+)]$ (see the inset, Fig. 3). The nonclassical nature of the field prevails despite the saturation effects that become important as Y approaches Y_{crit}. In Ref. [17], using a quantum stochastic method based on a large N expansion, Carmichael calculated a correlation function for a system when $x = 0.9$ with the same qualitative behavior as that of the one shown in the inset: antibunched and super-Poissonian.

Although there are limitations in the available theories, we have studied two different ones and found qualitative agreement in the evolution of the statistics as a function of intensity. The first is a calculation in the high-Q cavity limit (adiabatic elimination of the atoms) with a spatially varying field mode [18]. It shows a change from sub-Poissonian to super-Poissonian that remains in the presence of spatial mode functions, either a Gaussian or standing waves. Second, based on the plane wave theory of Ref. [19] without adiabatic elimination, but with an expansion in N, we have calculated $g^{(2)}(0)$ as a function of x and found that for a wide range of parameters the statistics of the light go from sub-Poissonian to super-Poissonian in a similar way as our observations. Other extensions that include transit broadening as a homogeneous process [20] and averaging over the number fluctuations [21] retain the qualitative agreement.

One place where a quantitative agreement with theory has been obtained is in measurements of the frequency of oscillation of $g^{(2)}(\tau)$. To concentrate on the frequency and not on the nonclassical effects in the amplitude, we operate the system in the intermediate regime of cavity QED, with $C_1 = 0.7, n_0 = 0.5$, and $N \approx 83$ (determined from the independent measurement of Ω_0). For these parameters and low driving intensity Eq. (1) predicts an oscillation of the correlation function at frequency Ω_0. Figure 4 shows the frequency of oscillation of $g^{(2)}(\tau)$ as a function of driving intensity Y. We extract the frequencies by the fast Fourier transform (FFT) of the data. We fit a Lorentzian to the FFT and take the center as the oscillating frequency Ω, assigning an uncertainty of the full width at half maximum of the peak divided by the signal to noise ratio of the transform. The normalized intensity axis has an overall uncertainty of ($\pm 10\%$) from the determination of Y_{crit}. In contrast with resonance fluorescence [22], where the Rabi frequency in the correlation function increases with excitation strength, Fig. 4 shows that in this cavity QED system increasing the driving intensity *decreases* the oscillation frequency of $g^{(2)}(\tau)$. This comes from the dependence

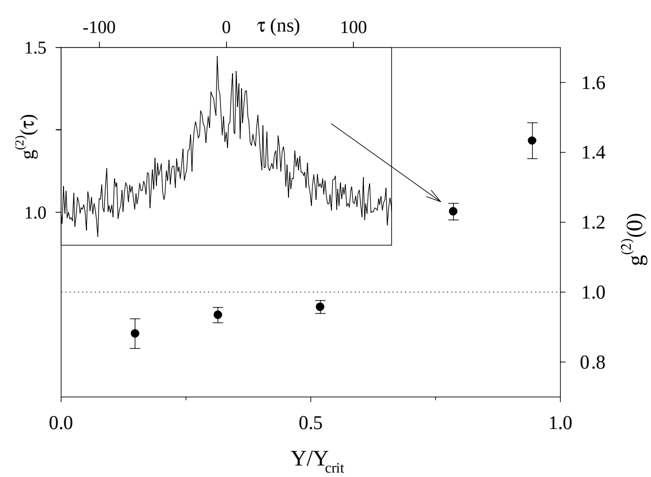

FIG. 3. Evolution of $g^{(2)}(0)$ as the intensity increases ($C_1 = 3.9$, $n_0 = 0.08$, and $N \approx 10$). The statistics change from sub-Poissonian to super-Poissonian. The inset presents the antibunched $g^{(2)}(\tau)$ for the point indicated in the plot.

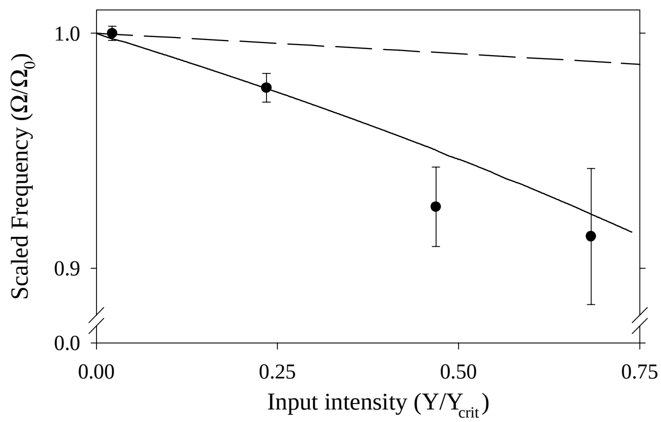

FIG. 4. Frequency of oscillation of $g^{(2)}(\tau)$ as a function of driving intensity $Y/Y_{\rm crit}$. The continuous curve is the prediction including the cavity geometry and the dashed curve is for a plane traveling wave mode ($C_1 = 0.7$, $n_0 = 0.5$, and $N \approx 83$).

of Ω on the intensity, e.g., in a plane traveling wave cavity through $g_0^2 \rightarrow g_0^2/(1 + |x|^2)$ [15]. The quantum regression theorem [19] establishes that the eigenvalues governing the time evolution of the correlation function are the same as those governing the time evolution of the expectation values. We find the linearized eigenvalues of the system away from the low intensity regime, including the standing waves and Gaussian transverse profile. We plot the frequency of the oscillatory part of the eigenvalue as the continuous curve in Fig. 4. The experimental measurements and the theory are in quantitative agreement. The dashed curve shows the result of a plane wave calculation which fails to reproduce our observations. We have observed this frequency change in the FFT of the time response to turn-on excitation [16].

This Letter shows measurements of the intensity correlation function of the light emitted by a system composed of N two-level atoms coupled to a single mode of the electromagnetic field. The escape of a photon from the cavity projects the system into a state where it evolves producing nonclassical correlations. We have generated a beam of light that violates the Schwarz inequality despite its bunched and super-Poissonian properties. The very large amplitude after $\tau = 0$ shows directly the effect of the projection of the wave function of the cavity QED system into a state with high polarization. The intrinsic delay of half a cycle to transfer the excitation back to the field is manifested in the correlation function as it returns to a steady state. The sub-Poissonian nature of the field at low driving intensities can change to super-Poissonian for increasing driving intensities, but the nonclassical nature remains with the presence of antibunching. We also have verified that the frequency of oscillation of the correlation function decreases with increasing input driving field showing the saturation induced decoupling between atoms and cavity.

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[1] E. T. Jaynes and F. W. Cummings, Proc. IEEE **51**, 89 (1963).
[2] R. J. Thompson, G. Rempe, and H. J. Kimble, Phys. Rev. Lett. **68**, 1132 (1992).
[3] G. Rempe, F. Schmidt-Kaler, and H. Walther, Phys. Rev. Lett. **64**, 2783 (1990).
[4] K. An, J. J. Childs, R. R. Dasari, and M. S. Feld, Phys. Rev. Lett. **73**, 3375 (1994).
[5] M. Brune, F. Schmidt-Kaler, A. Maali, J. Dreyer, E. Hagley, J. M. Raimond, and S. Haroche, Phys. Rev. Lett. **76**, 1800 (1996).
[6] D. M. Meekhof, C. Monroe, B. E. King, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **76**, 1796 (1996).
[7] G. Rempe, R. J. Thompson, R. J. Brecha, W. D. Lee, and H. J. Kimble, Phys. Rev. Lett. **67**, 1727 (1991).
[8] P. R. Rice and H. J. Carmichael, IEEE J. Quantum Electron. **24**, 1351 (1988).
[9] H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977).
[10] P. Grangier, G. Roger, A. Aspect, A. Heidmann, and S. Reynaud, Phys. Rev. Lett. **57**, 687 (1986).
[11] L. A. Lugiato in *Progress in Optics,* edited by E. Wolf (North-Holland, Amsterdam, 1984), Vol. XXI, pp. 69–216.
[12] *Cavity Quantum Electrodynamics,* edited by P. Berman Supplement 2 of Advances in Atomic, Molecular and Optical Physics Series (Academic Press, Boston, 1994).
[13] H. J. Carmichael, R. J. Brecha, and P. R. Rice, Opt. Commun. **82**, 73 (1991).
[14] A. L. Lacaita, F. Zappa, S. Bigliardi, and M. Manfredi, IEEE Trans. Electron Devices **40**, 577 (1993).
[15] J. Gripp, S. L. Mielke, L. A. Orozco, and H. J. Carmichael, Phys. Rev. A **54**, R3746 (1996); J. Gripp, S. L. Mielke, and L. A. Orozco, Phys. Rev. A **56**, 3262 (1997).
[16] S. L. Mielke, G. T. Foster, J. Gripp, and L. A. Orozco, Opt. Lett. **22**, 325 (1997).
[17] H. J. Carmichael, in *Frontiers in Quantum Optics,* edited by E. R. Pike and S. Sarkar (Hilger, Bristol, England, 1986).
[18] M. Xiao, H. J. Kimble, and H. J. Carmichael, Phys. Rev. A **35**, 3832 (1987).
[19] H. J. Carmichael, Phys. Rev. A **33**, 3262 (1986).
[20] R. J. Brecha, Ph.D. thesis, UT Austin, 1990; R. J. Brecha, P. Rice, and M. Xiao (to be published).
[21] G. Rempe, R. J. Thompson, R. J. Brecha, Q. Turchette, and H. J. Kimble, in Atomic Physics 13, edited by T. W. Hänsch, H. Walther, and B. Neizert, AIP Conf. Proc. No. 275 (AIP, New York, 1993).
[22] M. Dagenais and L. Mandel, Phys. Rev. A **18**, 2217 (1978).