Observation of Phase-sensitive Temporal Correlations in the Resonance Fluorescence from Two-Level Atoms

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Abstract

We have made the first observation of phase-dependent temporal correlations in the fluorescent field emitted by coherently driven two-level atoms in free space. We measure the temporal fluctuations of the fluorescent field when the resonant driving field is in-phase and out-of-phase, respectively, with the local oscillator field.

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Phase-sensitive detection of resonance fluorescence, i.e., allowing the light emitted from a coherently driven two-level atom in free space to interfere with a local oscillator (LO) beam, has received considerable attention throughout the history of quantum optics. In particular, phase-sensitive squeezing spectra for resonance fluorescence, first predicted more than 20 years ago [1], were recently observed for the first time [2]. Complementary to spectral measurement is the well-known use of homodyne or field autocorrelation techniques. Indeed, it is the temporal correlation function of the fluorescent field that yields direct information about the role of quantum jumps in phase-dependent resonance fluorescence. This point has been emphasized in the interpretation of recent spectral squeezing data obtained on coherently driven two-level atoms in free space [2, 3]. Further elegant elucidation of the role of quantum jumps in phase-dependent resonance fluorescence is provided by "conditional" homodyne detection [4, 5], where a measurement of the fluorescent field at $t+\tau$ is conditioned upon the unequivocal determination that the atom made a quantum jump to the ground state at an earlier time t [6]. Hence, it is clear that a direct measurement of the phase-sensitive two-time field autocorrelation function for the fluorescent field is of considerable interest.

In this Letter, we report the first observation of temporal correlations in the phase-dependent fluorescence of a beam of driven two-level atoms *in free space*. Our data agree well with a rigorous theoretical expression for the two-time autocorrelation function of the field emitted by a single two-level atom that was published earlier [7]. We measure the temporal fluctuations of the fluorescent field in the two cases where the coherent driving field is in-phase, and out-of-phase, with the LO. Hence we measure the fluctuations in the induced atomic dipole that are in-phase and out-of-phase, respectively, with the driving field.

To measure single-atom phase-dependent effects in resonance fluorescence, the fluorescent field $\hat{E}_{fl}(t)$ from a coherently driven atom is mixed with a local oscillator (LO) field $|E_{LO}|e^{i\phi}$ having a controllable fixed phase ϕ relative to the driving field [2, 7]. The two-time correlation function of the resultant field is then measured by homodyne detection. Because the LO is strong compared to the fluorescent field, the dominant atomic contribution comes from the interference term between the radiated field and the LO. The field radiated by the atom thus causes fluctuations in the detected power $\Delta\hat{P}$ given by

$$\Delta\hat{P}(t) \propto |E_{LO}|(e^{-i\phi}\hat{E}_{fl}(t) + e^{i\phi}\hat{E}_{fl}^{\dagger}(t)) \tag{1}$$

where the caret denotes a quantum operator. When the relative phase ϕ is set to 0^0 (90^0), the LO is in-phase (out-of-phase) with the driving field, and fluorescent field fluctuations in-phase (out-of-phase) with the driving field are detected. Using standard notation $\hat{\sigma}_{+/-}(t)$ for the atomic dipole raising / lowering operators of a two-level atom, we obtain $\hat{E}_{fl}(t) \propto \hat{\sigma}_{+}(t)$ and $\hat{E}_{fl}^{\dagger}(t) \propto \hat{\sigma}_{-}(t)$, where retardation effects from the atom to the detector are neglected. These fluctuations are measured with a fast photodiode and the normalized correlation function $C(\tau)$ is then determined by

$$C(\tau) = \langle: \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) :\rangle_t / \langle\left(\Delta\hat{P}(t)\right)^2\rangle_t. \tag{2}$$

Here $\langle\ldots\rangle$ denotes an ensemble average and the subscript "t" denotes that an averaging over the initial times t is performed.

Our experimental setup has been described in detail earlier [2, 7]. We briefly outline the salient points here. A supersonic beam of two-level atoms is coherently driven by cw laser beams propagating at right angles to the atomic beam. In order to observe phase-sensitive quantum fluctuations with high signal-to-noise ratio [2, 9], we employ a novel homodyne detection scheme that measures the radiation scattered by the atoms only along the path of propagation of the driving field [8]. In this scheme, orthogonal polarizations of the same laser beam are used to create mode-matched LO and driving fields between which a well-defined controllable relative phase ϕ is inserted with a Babinet compensator. We set $\phi = 0^0$ to measure the fluorescent fluctuations that are in-phase with the driving field, and $\phi = 90^0$ to measure the fluctuations that are out-of-phase. Further, transmitted power signals through *two* identically prepared atomic samples are obtained using diode detectors, and then subtracted (see Fig. 1). This accomplishes high suppression of technical noise in *both* the LO and the quadrature signals, an improvement over ordinary homodyne detection with a beamsplitter [7].

After subtraction of the diode signals, the difference photocurrent is converted to a voltage, then AC coupled and further amplified. The resulting time waveforms $V(t)$ are transferred to a computer which calculates the normalized correlation $C(\tau) = \langle V(t)V(t + \tau)\rangle_t / \langle V(t)^2\rangle_t$. Note that simply measuring the voltage waveforms emanating from our subtraction electronics yields no discernable information, as shown in the voltage-time plot in Fig. 2. However, examining how the fluctuations at different instants within this waveform are correlated, does reveal interesting information as described below.

We start by gating the atomic beam off to measure our "noise floor". In this case we simply measure shot noise. As shown in the inset in Fig. 3, the autocorrelation function $C_{\text{shot}}(\tau)$ of shot noise is zero at long times and has a sharp peak near $t = 0$, as expected. This peak has finite width because of the finite bandwidth (20 MHz) of the amplifier. The initial dip below zero is because the signal is AC-coupled, hence the area under the curve must be zero. Next, the atomic beam is turned on and phase-dependent correlations in fluorescence $C_\phi(\tau)$ are measured. The shot noise correlation function $C_{\text{shot}}(\tau)$ is subtracted from $C_\phi(\tau)$, to finally obtain $C(\tau) = C_\phi(\tau) - C_{\text{shot}}(\tau)$. The total power P incident on the detectors, as measured with a power meter, is reset to the same value with the atomic beam turned on or off. Plots [a] and [b] in Fig. 3 show the measured in-phase ($\phi = 0^0$) and out-of-phase (90^0) autocorrelation function for on-resonance excitation ($\Delta = 0$). Each point on the plot is an average over 150,000 data points accumulated over ten minutes. The overall magnitude of the detected signal is much smaller when the driving and LO fields are out-of-phase with each other, than when they are in-phase. This degrades the quality of the out-of-phase data causing a slight dip at small times in Fig. 3(b) when we subtract $C_{\text{shot}}(\tau)$ from $C_{\phi=90^0}(\tau)$.

We now examine how well our data compares with the results of an earlier rigorous quantum calculation of the autocorrelation function based on operator Bloch vector equations [7]. Relaxation effects owing to spontaneous emission may be neglected because the two-level atom in our experiment consists of the ${}^1S_0 \rightarrow {}^3P_1$ 556 nm transition of ^{174}Yb. The radiative lifetime of this transition is 875 ns, long compared to the laser-atom interaction time τ_0 for our apparatus, where τ_0 is the transit time of the atoms through the laser field. Each laser beam in Fig. 1 is focused to a $1/e$ field radius of 0.13 mm along the atomic beam, yielding $\tau_0 \approx 372$ ns [2]. These long-lived two-level atoms are strongly-driven [2, 3]: The driving field power is 2 mW (one region) yielding a Rabi frequency of 5.9 MHz. The LO power is 1.1 mW.

In the dipole approximation, the field radiated by the atom $\hat{E}_{fl}(t)$, as measured at some spatial location, is proportional to the light-induced atomic dipole at that instant, neglecting retardation effects. From Ref. [3, 7] we obtain using Eqns. 1 and 2:

$$C(\tau) = (1 - \tau/\tau_0)\left[\frac{\Delta}{2\Omega'}\sin 2\phi \sin \Omega'\tau + \cos^2\phi \cos \Omega'\tau + \sin^2\phi\right] \tag{3}$$

where the dependencies on the in-phase fluctuations, the out-of-phase fluctuations, and the cross-correlations between the in- and out-of-phase fluctuations are clearly displayed. Here

$\Omega' = \sqrt{\Omega^2 + \Delta^2}$ where Ω is the Rabi frequency and Δ is the laser detuning, and a common factor proportional to $\frac{\Omega^2}{4\Omega'^2}|E_{LO}|^2$ has been suppressed. Note that the pre-factor $1 - \tau/\tau_0$ in Eqn. 3 results from the average over initial times t $(0 \leq t \leq \tau_0 - \tau,$ where $0 \leq \tau \leq \tau_0)$ where we use the fact that we are in the strong driving limit $\Omega'\tau_0 \gg 1$. This transit-time pre-factor clearly arises from the fact that the probability of detecting two photons τ apart, from a passing atom, has to decrease as τ approaches the transit time τ_0.

For on-resonance excitation ($\Delta = 0$) the cross-correlations between the in- and out-of-phase fluctuations, contained in the sin2ϕ term, disappear. In this case, if we set $\phi = 0^0$ in order to extract the fluctuations of the fluorescent field in-phase with the driving field, we obtain just the $\cos^2$ term which exhibits a cosine oscillation at the Rabi frequency Ω (the amplitude of this oscillation decreases with increasing τ owing to the pre-factor $1 - \tau/\tau_0$). This theoretical prediction is plotted (dashed line) in Fig. 3(a). On the other hand, if we set $\phi = 90^0$ in order to extract the fluctuations of the fluorescent field out-of-phase with the on-resonance driving field, we obtain just the $\sin^2$ term which is independent of τ (the only dependence on τ is through the trivial transit-time pre-factor $1 - \tau/\tau_0$). This theoretical prediction is plotted (dashed line) in Fig. 3(b).

The data in Fig. 3 agrees well with the theory, and can be simply explained. Note that only the dipole component that radiates in-phase with the LO is actually detected. When the LO is in-phase with the driving field ($\phi = 0^0$), the detected component of the mean atomic dipole radiates in-phase with the driving field, hence oscillations in $C(\tau)$ at the Rabi frequency are expected, and observed. When the LO is out-of-phase with the driving field ($\phi = 90^0$), one may initially expect that the driven atomic dipole would radiate out-of-phase with the LO and hence not be detected. However, owing to the random phase nature of spontaneous emission there is a constant 50% likelihood of radiating in-phase with the LO, which leads to a non-zero detectable component of the dipole that radiates out-of-phase with the driving field. This would ideally result in a flat line at unity for $C(\tau)$. However, $C(\tau)$ actually slopes downward in Fig. 3(b) because of the trivial transit-time dependence as mentioned in the previous paragraph.

In conclusion, we have measured and theoretically explained temporal correlations in resonance fluorescence for the two cases where the resonant driving field is in- and out-of-phase respectively with the LO. As a possible future direction it would be interesting to examine the case of nonzero detuning; then the sin2ϕ term in Eqn. 3 does not vanish. This term describes a cross-correlation between the in- and out-of-phase fluctuations and has been shown to be responsible for all phase-sensitive squeezing effects in resonance fluorescence [2]. In Lu, et al, maximum squeezing was observed near the $\phi = \pm 45^0$ quadratures for strong driving fields and nonzero detuning. From Eqn. 3, we see that $C_{\pm 45^0}(\tau) = \frac{1}{2}(1 - \tau/\tau_0)\left[1 + \cos\Omega'\tau \pm \frac{\Delta}{\Omega'}\sin\Omega'\tau\right]$. This implies that in the vicinity of small values of τ one would observe antibunching in one quadrature and bunching in the other.

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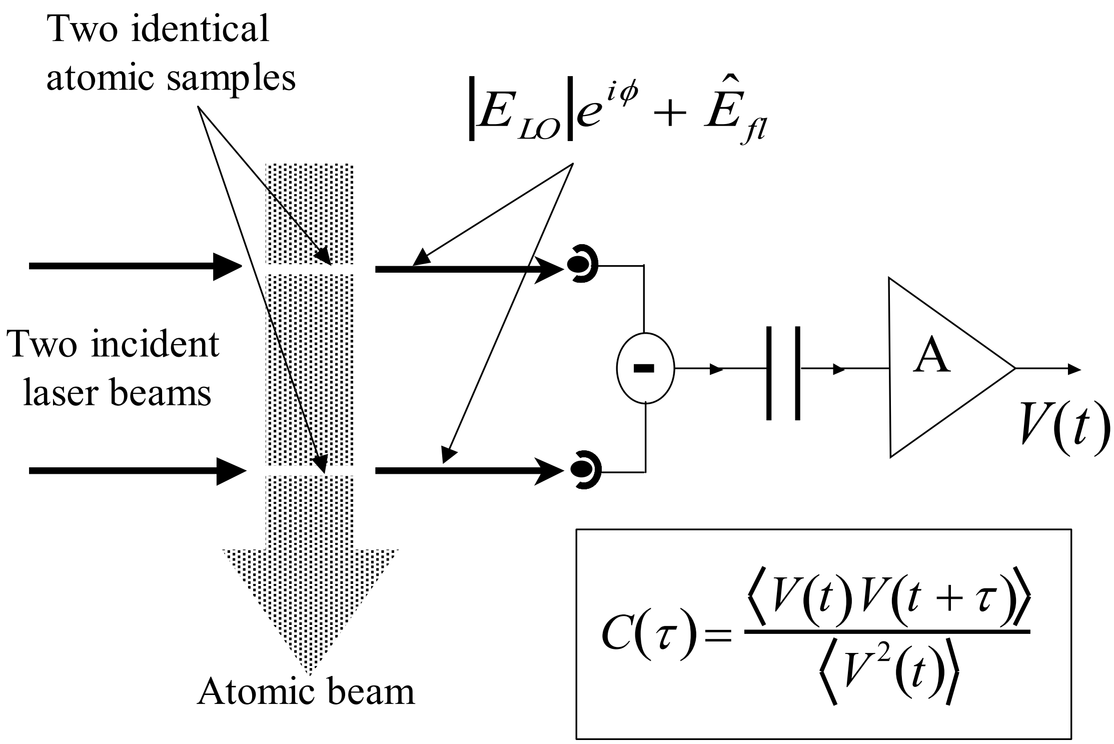

Fig.1, Lu, et al.

FIG. 1: Outline of measurement of phase-dependent temporal correlations in resonance fluorescence by subtraction of transmitted power signals from two identically prepared atomic samples. The transit time τ_0 is the time taken by the atoms to pass through each of the two laser beams.

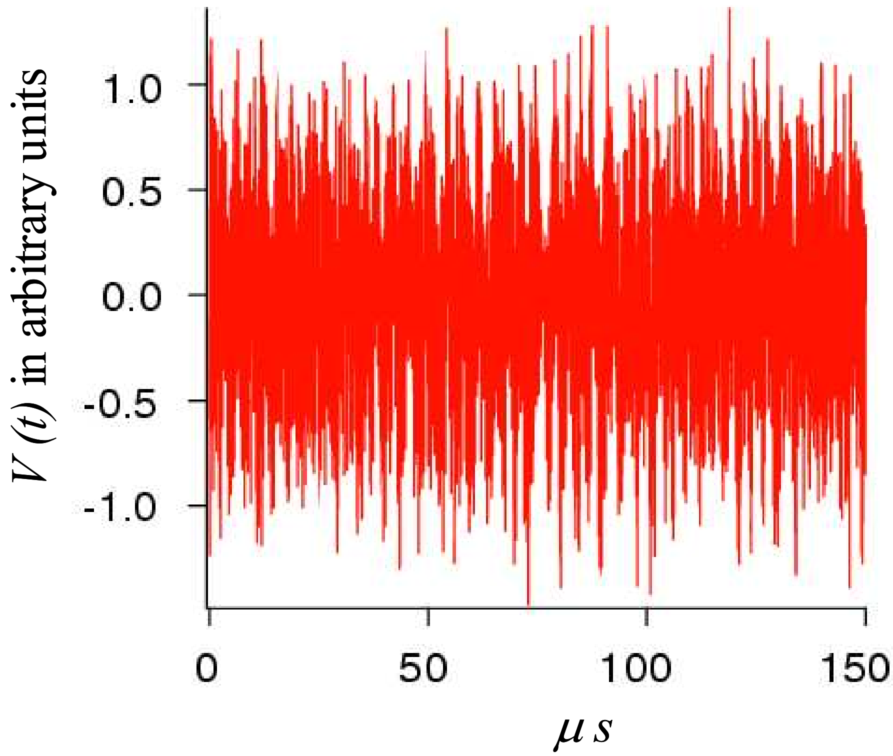

Fig. 2 Lu, et al.

FIG. 2: The voltage waveform *V(t)* emanating from the subtraction electronics yields no discernable information.

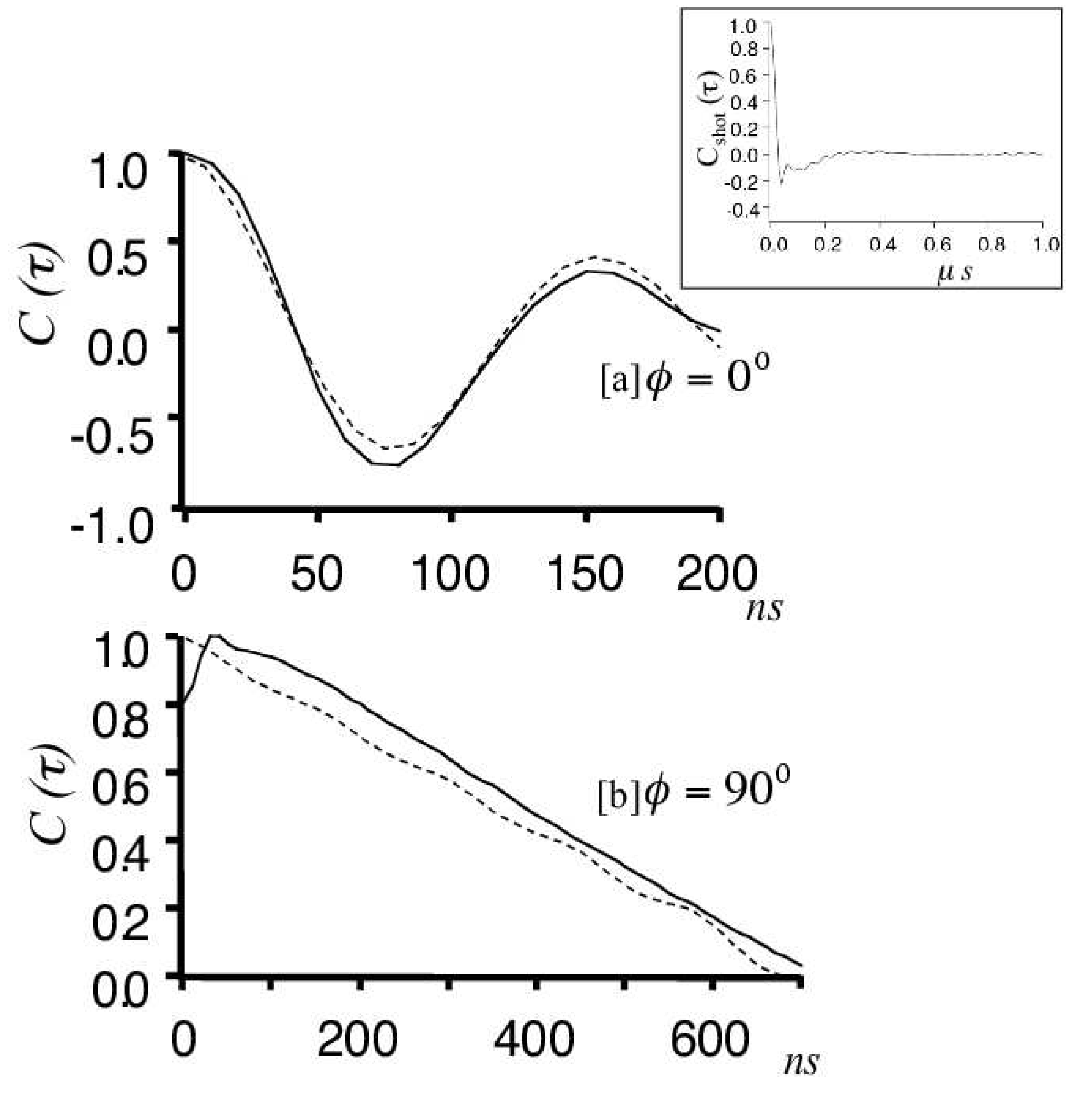

Fig. 3, Lu et al.

FIG. 3: [a] In-phase (i.e., $\phi = 0^0$) and [b] out-of-phase (i.e., $\phi = 90^0$) two-time autocorrelation function $C(\tau)$ as a function of delay τ for on-resonance excitation ($\Delta = 0$). The solid lines are data. The dashed lines are not best-fits, they are obtained from Eqn. 3 using the experimental parameters given in the text. The inset shows the autocorrelation $C_{\text{shot}}(\tau)$ for the shot noise, i.e., when the atomic beam is gated off.