Physical Interpretation of Phase-Dependent Resonance Fluorescence

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ABSTRACT

We investigate the phase properties of the fluorescent field emitted by two-level atoms which are coherently driven by a near-resonant laser field in free space. The phase-sensitive two-time field correlation function is constructed using simple physical arguments, and two distinct contributions are identified: one corresponding to atomic emission from a partly excited state, and the other from the fully excited state. Measurements of the phase-sensitive temporal correlations are presented that, along with our calculations, elucidate the difference between the fluorescent field emitted from a partially and a fully excited atom. Further, we show that squeezing arises when a *partially* excited atom makes a quantum jump to the ground state, and that this same physical process causes squeezing in *both* short-lived and long-lived atoms.

Keywords: resonance fluorescence, squeezing, temporal correlation

1. INTRODUCTION

Phase-sensitive detection of resonance fluorescence, i.e., allowing the light emitted from an atom undergoing Rabi oscillations in free space to interfere with a coherent 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. Our calculation of the phase-sensitive temporal correlations suggests interesting physical insights into the role played by quantum jumps. Therefore, a direct measurement of the two-time field autocorrelation function for the fluorescent field is of considerable interest. Here we report measurements of temporal correlations in the phase-dependent fluorescence of a beam of strongly driven two-level atoms in free space. Our data agrees well with a theoretical expression for the two-time autocorrelation function of the field emitted by a single two-level atom that was published earlier.[2] Using simple physical arguments we elucidate the difference between the phase properties of fluorescence from a partly excited and a fully excited atom.

The second aim of our paper is to point out that phase-sensitive squeezing[1] in resonance fluorescence is a well-studied topic in theory,[3] but is relatively less explored in experiment. On the one hand, traditional theoretical treatments of phase-sensitive squeezing have focused on the case of weak, on-resonant excitation, and emphasized homodyne detection of the in- and out-of-phase (0^0 and 90^0 respectively) quadratures. On the other hand, the only observation to date of squeezing spectra[2] in resonance fluorescence was made at strong excitation with non-zero detuning, via homodyne detection at a phase near $\pm 45^0$ relative to the driving field. Further, past theoretical predictions[1] were made in the context of a "short-lived" atom, where the observation time for laser-atom interaction was much greater than the natural atomic lifetime, meaning that relaxation effects dominate. But the experiment was performed on a "long-lived" atom, where the atomic lifetime far exceeded the interaction time, meaning that relaxation effects can be ignored. Here, we show that despite the seeming disparateness between theory and experiment, the squeezing predicted previously for short-lived atoms as well as the squeezing observed for long-lived atoms is caused by the same physical process: a quantum jump by a *partially* excited atom to the ground state.

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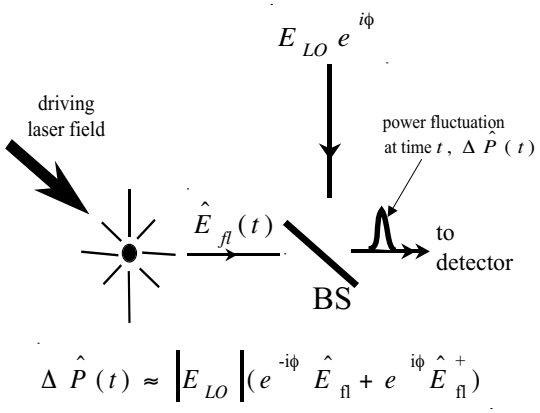

Figure 1. Measurement of phase-dependent effects in resonance fluorescence by homodyne detection. Fluorescence from a resonantly driven atom is mixed with a local oscillator field (LO) at a beamsplitter (BS). The LO is derived from the driving laser field.

2. PHASE-DEPENDENT RESONANCE FLUORESCENCE

Resonance fluorescence is a particularly well-explored topic in quantum optics.[4,5] *Phase-dependent* effects in resonance fluorescence were measured[2,6,7] by mixing the fluorescent field $\hat{E}_{fl}(t)$ (the caret denotes a quantum operator) with a local oscillator (LO) field $|E_{LO}|e^{i\phi}$ having a controllable fixed phase ϕ relative to the driving field (Fig. 1). The interference between the atom field and the LO causes a resultant field $\Delta\hat{P}$ where

$$\begin{aligned}\Delta\hat{P}(t) &= |E_{LO}|(e^{-i\phi}\hat{E}_{fl}(t) + e^{i\phi}\hat{E}^{\dagger}_{fl}(t)) \\ &= \vec{K}(\vec{r})|E_{LO}|\left[e^{-i\phi}\hat{\sigma}_{+}(t) + e^{i\phi}\hat{\sigma}_{-}(t)\right]. \end{aligned} \quad (1)$$

Here we've used $\hat{E}_{fl}(t) = \vec{K}(\vec{r})\hat{\sigma}_{+}(t)$ where $\vec{K}(\vec{r}) = \frac{\omega^2}{4\pi\epsilon_0 c^2}\left(\frac{\vec{d}}{r} - \frac{(\vec{d}.\vec{r})\vec{r}}{r^3}\right)$ is the usual spatial dipole pattern at point $\vec{r}$ radiated by an electric dipole $\vec{d}$ oscillating at frequency ω, and $\hat{\sigma}_{\pm}(t)$ are standard notation for the atomic dipole raising and lowering operators.[4,5]

The two properties of the resultant field we're interested in are a) the two-time phase-sensitive correlation function $\langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle$, and b) its Fourier transform the phase-dependent power spectrum, which is intimately related to the squeezing spectrum $S_{\phi}(\omega)$.[3] We investigate these two quantities in the following sections.

3. THE PHASE-SENSITIVE TWO-TIME AUTOCORRELATION FUNCTION

Our experimental setup has been described in detail earlier.[2,7] We use a continuous laser field to strongly drive the $^1S_0 \rightarrow {}^3P_1$ 556 nm transition of ^{174}Yb. The radiative lifetime for this transition is 875 ns, long compared to the interaction time in our apparatus. A novel homodyne detection scheme, that relies upon observing the radiation scattered by the atoms only in the forward direction, i.e., along the path of propagation of the driving field (Fig. 2), is used to measure phase-sensitive quantum fluctuations with high signal-to-noise ratio.[2] 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 $\phi = 0^0$ (90^0) to measure fluorescent fluctuations in-phase (out-of-phase) with the driving field. Further, transmitted power signals through *two* identically prepared atomic samples are obtained, and then subtracted which accomplishes high suppression of technical noise in *both* the LO and the quadrature signals,

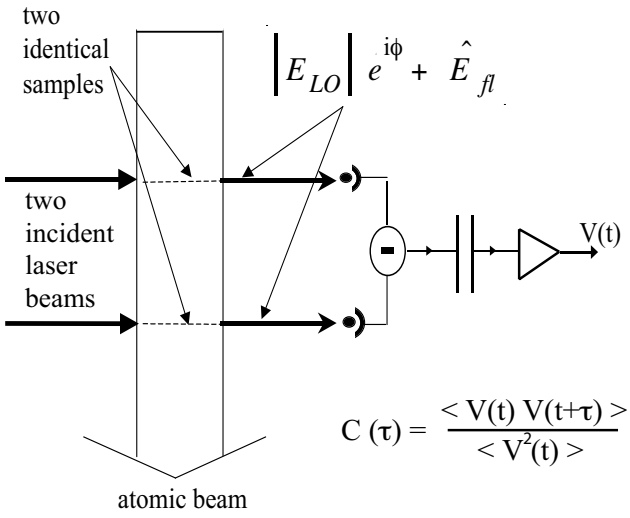

Figure 2. Outline of measurement of phase-dependent temporal correlations in resonance fluorescence by subtraction of transmitted power signals from two identically prepared atomic samples.

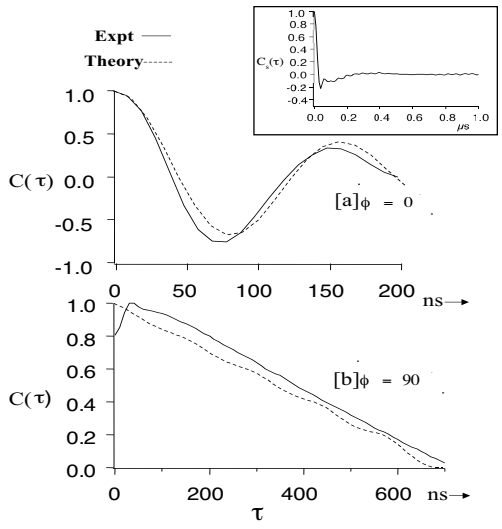

Figure 3. [a] In-phase (i.e., $\phi = 0^0$) and [b] out-of-phase (i.e., $\phi = 90^0$) two-time autocorrelation function $C(\tau)$ as a function of delay τ for on-resonance excitation ($\Delta = 0$). The inset shows the autocorrelation $C_S(\tau)$for the shot noise.

better than ordinary homodyne detection with a beamsplitter. After subtraction the photocurrent is converted to a voltage, then AC coupled and further amplified. The resulting time waveforms $V(t)$ are scanned into a digital oscilloscope and transferred in real time through a GPIB cable to a computer where the normalized correlation $C(t, t+\tau) = \langle V(t)V(t+\tau)\rangle / \langle V^2\rangle$ is formed. After averaging over different initial times t, $\langle C(t, t+\tau)\rangle_t = C(\tau)$, τ being the time interval between two consecutive photodetections. Note that for a long-lived atom, if two photons are detected a time τ apart they are successively emitted photons, i.e., the probability of the atom radiating between t and $t+\tau$ is negligible.

Fig. 3 shows the measured in-phase ($\phi = 0^0$) and out-of-phase (90^0) autocorrelation function for on-resonance excitation ($\Delta = 0$). The laser fields are focused to a $1/e$ field radius of 0.13 mm along the atomic beam, and 1 mm in the orthogonal direction, as measured with a 0.025 mm pixel diode array. To measure our "noise floor"

we gate the atomic beam off and measure the autocorrelation function $C_S(\tau)$ of shot noise (inset, Fig. 3). As expected, $C_S(\tau)$ is zero at long times and has a sharp peak near $t = 0$ with 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 $C_S(\tau)$ is subtracted from $C_\phi(\tau)$, to finally obtain $C(\tau) = C_\phi(\tau) - C_S(\tau)$. We make sure that 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.

The measured in-phase correlation function basically oscillates at the Rabi frequency. On the other hand the out-of-phase autocorrelation is essentially a line that slopes downward as the delay increases. The slight dip at small times in the $\phi = 90^0$ experimental curve in Fig. 3 occurs because 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 dip when we subtract $C_S(\tau)$ from $C_{\phi=90^0}(\tau)$.

To understand the data in Fig. 3 we now calculate the phase-sensitive two-time autocorrelation function $\langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle$ for the conditions of the experiment, i.e., for strong off-resonant excitation of long-lived atoms. This calculation uses operator Bloch vector equations[7] with nonzero detuning but neglects spontaneous emission because the radiative lifetime of the long-lived atom far exceeded the laser-atom interaction time τ_0. The calculation of the two-time autocorrelation function involves the evaluation of the expectation of the two-time correlation functions $\hat{\sigma}_+(t)\hat{\sigma}_\pm(t+\tau)$ and $\hat{\sigma}_\pm(t+\tau)\hat{\sigma}_-(t)$. The result[7] is given below:

$$\begin{aligned}\langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle &= \cos^2\phi\,[\sin\Omega' t \sin\Omega'\tau + (1-\cos\Omega' t)\cos\Omega'\tau] \\ &+ \sin^2\phi(1-\cos\Omega' t) \\ &+ \frac{\Delta}{\Omega'}\sin\phi\cos\phi\,[\sin\Omega' t(1-\cos\Omega'\tau) + (1-\cos\Omega' t)\sin\Omega'\tau]\,. \end{aligned} \quad (2)$$

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.

Similarly, in order to derive $\langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle$ for short-lived atoms we again start with the optical Bloch equations, but include relaxation.[3–5] The relaxation is denoted by $1/\gamma$, the $1/e$-decay time with which the atomic excited state decays exponentially in time. The interaction time far exceeds the relaxation time, i.e., $\tau_0 \gg 1/\gamma$. We set the detuning equal to zero because maximum squeezing in this case occurs for on-resonant excitation.[1,3] Note that past theoretical treatments omitted to clearly articulate a role for the LO, opting instead to calculate phase-dependent fluorescent fluctuations in terms of a relative phase between the radiated field and the driving field, denoted by angle θ.[1,3] This means that θ is different from ϕ (defined above as the relative angle between the LO and the driving field). However, the role of θ and ϕ is the same: Fluorescent fluctuations in-phase (out-of-phase) with the driving field are calculated by setting $\theta = 0\,(90^0)$. After solving the coupled Bloch equations for the mean dipole and the mean inversion we use the quantum regression theorem to calculate the same two-time correlation functions for $\sigma_\pm$ as mentioned above for the case of the long-lived atom. These two-time correlation functions are well-known.[5] In analogy to Eqn. 2 we obtain,

$$\langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle = \left[\{a_1(t)a_1(\tau) + b_1(t)a_3(\tau)\}\,e^{-2i\theta} + a_1(t)a_1^*(\tau) + b_1(t)a_2(\tau)\right] + c.c., \quad (3)$$

where the a and b coefficients are given in Ref.[5] as:

$$\begin{aligned} a_1(t) &= -i\frac{\Omega\gamma}{\gamma^2+2\Omega^2}\left[1 - e^{-3\gamma t/4}\left(\cos\mu t - (\frac{4\Omega^2-\gamma^2}{4\gamma\mu})\sin\mu t\right)\right] \\ a_{2,3}(t) &= \frac{1}{2}e^{-\gamma t/2} \pm \frac{1}{8\mu}e^{-3\gamma t/4}\,(\gamma\sin\mu t + 4\mu\cos\mu t) \\ b_1(t) &= \frac{\Omega^2}{\gamma^2+2\Omega^2}\left[1 - e^{-3\gamma t/4}\left(\cos\mu t + \frac{3\gamma}{\mu}\sin\mu t\right)\right], \end{aligned} \quad (4)$$

where $\mu = \sqrt{\Omega^2 - \gamma^2/16}$. Note that for weak excitation such that $\Omega < \gamma/4$, Eqns. 4 are expressed in terms of non-oscillatory hyperbolic functions instead of trigonometric.

In order to track down the physical process responsible for squeezing, and also gain other interesting physical insights into phase-dependent resonance fluorescence, it is far more profitable to re-express the above results for $C(t,t+\tau) = \langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle$ as a sum of two contributions as follows: For a two-level atom illuminated by a near-resonant coherent exciting field, the expectation of the atomic excitation changes continuously at the Rabi frequency. The atom may emit spontaneously when the atom is in a superposition of the ground and the excited state (i.e., partially excited "p.e."), or when the atom is in the upper energy eigenstate (i.e., fully excited "f.e."). Both possibilities contribute to $C(t,t+\tau)$, hence

$$\begin{aligned} \langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle = C(t,t+\tau) &= C_{p.e.}(t,t+\tau) + C_{f.e.}(t,t+\tau) \\ &= \langle\Delta\hat{P}(t)\rangle\langle\Delta\hat{P}(\tau)\rangle_g + \frac{1}{2}P_e(t)M(\tau), \end{aligned} \tag{5}$$

where $C_{p.e.}(t,t+\tau) = \langle\Delta\hat{P}(t)\rangle\langle\Delta\hat{P}(\tau)\rangle_g$ and $C_{f.e.}(t,t+\tau) = \frac{1}{2}P_e(t)M(\tau)$. Equation 5 is true for both long-lived as well as short-lived atoms - of course, the symbols P_e, $\Delta\hat{P}$, and M stand for different expressions in either case. This equation is the cornerstone of our physical interpretation of phase-dependent resonance fluorescence. Let us now examine closely the meaning and implications of the symbols in Equation 5.

The first term on the right hand side of Eqn. 5 describes the joint probability of photodetection when the fluorescent field from a *partly* excited atom (i.e.,in some superposition of the ground and excited states) is mixed with the LO. The probability of the first photodetection occurring at time t corresponds to $\langle\Delta\hat{P}(t)\rangle = |E_{LO}|(e^{-i\phi}\langle\hat{E}_{fl}(t)\rangle + e^{i\phi}\langle\hat{E}^{\dagger}_{fl}(t)\rangle)$ from Eqn. 6. For a strongly driven long-lived atom we obtain,[2]

$$\langle\Delta\hat{P}(t)\rangle = -\left(\frac{\Omega}{\Omega'}\frac{\sin\Omega' t}{2}\cos\phi + \frac{\Omega\Delta}{\Omega'^2}\frac{1-\cos\Omega' t}{2}\sin\phi\right), \tag{6}$$

while for a weakly-driven short-lived atom we obtain

$$\langle\Delta P(t)\rangle = -i\frac{\Omega\gamma}{\gamma^2+2\Omega^2}\left[1 - e^{-3\gamma t/4}\left(\cos\mu t - (\frac{4\Omega^2-\gamma^2}{4\gamma\mu})\sin\mu t\right)\right]e^{-i\theta} + c.c. \tag{7}$$

Note that in the context of our data (at $\Delta = 0$) in Fig. 3, there is no contribution to $C(\tau)$ from $C_{p.e.}(\tau)$ as explained below. The probability of the first photodetection occurring at time t corresponds to $\langle\Delta\hat{P}(t)\rangle = |E_{LO}|(e^{-i\phi}\langle\hat{E}_{fl}(t)\rangle + e^{i\phi}\langle\hat{E}^{\dagger}_{fl}(t)\rangle)$. In our case, for long-lived atoms and strong excitation, we obtain[2] $\langle\Delta\hat{P}(t)\rangle = -\vec{K}(\vec{r})\left(\frac{\Omega_R}{\Omega'}\frac{\sin\Omega' t}{2}\cos\phi - \frac{\Omega_R\Delta}{\Omega'^2}\frac{1-\cos\Omega' t}{2}\sin\phi\right)$, where Ω_R is the Rabi frequency and the generalized Rabi frequency $\Omega' = \sqrt{\Omega^2+\Delta^2}$. $\vec{K}(\vec{r})$ denotes the usual E-field radiated by an oscillating dipole.[4] A key feature of a radiating quantum oscillator is that immediately following a detection at time t the atom wavefunction *must collapse to the ground state*. Hence the next detection $\langle\Delta\hat{P}(t+\tau)\rangle$ at time $t+\tau$ depends only on the time difference τ, i.e., $\langle\Delta\hat{P}(t+\tau)\rangle_{g@t} = \langle\Delta\hat{P}(\tau)\rangle$. For on-resonant excitation we see that $\langle\Delta\hat{P}(t)\rangle \propto \sin\Omega_R t\,\cos\phi$, which goes to zero when averaged over different initial times t, meaning that $C_{p.e.}(\tau) \simeq 0$.

The second term on the right hand side of Eqn. 5 describes the joint probability of photodetection when the spontaneous emission from a *fully* excited atom (i.e., in the upper energy eigenstate) is mixed with the LO. $P_e(t)$ is the probability of the atom being in the upper energy eigenstate [2,7] at time t. For a strongly-driven long-lived two-level atom initially in the ground state

$$P_e(t) = \frac{\Omega^2}{2\Omega'^2}(1 - cos\Omega t), \tag{8}$$

while for a weakly-driven short-lived atom initially in the ground state,

$$P_e(t) = \frac{\Omega^2}{\gamma^2+2\Omega^2}\left[1 - e^{-3\gamma t/4}\left(\cos\mu t + \frac{3\gamma}{\mu}\sin\mu t\right)\right]. \tag{9}$$

The reason for the pre-multiplier 1/2 in Eqn. 5 is as follows: Because the atom emits a pure single-photon Fock state from the upper energy eigenstate the optical phase is completely random[8] (since the number and

phase operators do not commute). Therefore, this emission can be resolved into two equal components - one in-phase with the LO which is detected with 50% likelihood, the other out-of-phase with the LO which is never detected.

The next factor in the first term of Eqn. 5 is $M(\tau)$ which describes the evolution of the mean atomic dipole at times τ *after* the first photodetection at time t.[2] For strongly-driven long-lived atoms we find $M(\tau) = \cos^2\phi\ \cos\Omega'\tau + \sin^2\phi\left[\frac{\Omega^2}{\Omega'^2} + \frac{\Delta^2}{\Omega'^2}\cos\Omega'\tau\right]$. For weakly-driven short-lived atoms we find that $M(\tau) = 4\left[e^{-\gamma\tau/2}\cos^2\theta - \frac{e^{-3\gamma\tau/4}}{4\mu}(\gamma\sin\mu\tau - 4\mu\cos\mu\tau)\sin^2\theta\right]$. Note that the on-resonant data in Fig. 3 is completely dominated by the contribution from the fully excited state $C_{f.e.}(t, t+\tau)$ and, as a result of averaging over t in the data, is really a map of $M(\tau)$. A simple understanding of the above expression for $M(\tau)$ is possible for the case of zero detuning, $\Delta = 0$, which is indeed the condition in which our data in Fig. 3 has been taken. 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 ($\phi = 0$), the detected component of the mean atomic dipole radiates in-phase with the driving field, hence oscillations at the Rabi frequency are predicted[2, 6, 7] by the above expression for $M(\tau)$. These oscillations are indeed observed in Fig. 3 (a). When the LO is out-of-phase with the driving field ($\phi = 90$), one may be tempted to make the naive prediction that the driven atomic dipole radiates completely out-of-phase with the LO and hence is not detected. However, owing to the random phase nature of spontaneous emission, there is a constant component of the mean atomic dipole that is out-of-phase with the driving field, therefore in-phase with the LO. This would ideally lead to a flat line at unity for $C(\tau)$. However, the data in Fig. 3 (b) exhibits a downward slope because the laser-atom interaction time is limited by the transit time τ_0 of the atom through the laser beam. In an extreme situation, the probability of detecting two photons τ apart falls to zero if t is such that $\tau_0 - t < \tau$. Further explanation, including a discussion of non-zero detuning, is deferred to a forthcoming publication.[6]

Thus we see that Eqn. 5 has enabled an important physical insight into the process of phase-dependent resonance fluorescence, which was being obscured by the math in Eqns. 2, 3, and 4. This insight is true no matter whether we're dealing with off-resonantly excited long-lived atoms where relaxation is ignored, or with resonantly excited short-lived atoms where the relaxation γ needs to be included. This physical insight is as follows: A coherently excited atom can either spontaneously emit from the fully excited state or from the partly excited state. Upon combining this spontaneous emission with the LO field one obtains the two-time autocorrelation $\langle : \Delta\hat{P}(t)\Delta\hat{P}(t+\tau) : \rangle$ which neatly separates out into two terms that reflect either possibility. In particular, the term describing phase-sensitive two-time detection of spontaneous emission from a partly excited atom shows a quantum collapse of the atomic wavefunction to the ground state immediately following the first photodetection.

In the next section we use this insight provided by Eqn. 5 to show that is this quantum collapse to the ground state that leads to squeezing in phase-dependent resonance fluorescence.

4. THE SQUEEZING SPECTRUM

4.1. The Short-Lived Atom

We begin with the case of short-lived atoms because it is in their context that the squeezing spectrum $S_\phi(\omega)$ has been described previously in the literature. The usual definition of the squeezing spectrum is given in Ref.[3] as

$$S_\phi(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \langle : \Delta\hat{P}(t), \Delta\hat{P}(t+\tau) : \rangle e^{i\omega\tau}, \tag{10}$$

where $\langle \hat{A}, \hat{B} \rangle \equiv \langle \hat{A}\hat{B} \rangle - \langle \hat{A} \rangle \langle \hat{B} \rangle$, and the $\Delta\hat{P}$ operators are phase-dependent (Eqn. **??**). Because of the inclusion of relaxation any transient oscillations damp out soon and the mean intensity radiated by the fluorescing atom settles into a "steady state" quickly. If the interaction time includes many relaxation cycles, i.e. $\tau_0 \gg 1/\gamma$, then it is appropriate to evaluate the two-time correlation function in Eqn. 10 in the "long-time limit" $t \to \infty$. Therefore, inserting $t \to \infty$ in Eqn. 5 and using the definition of $\langle \hat{A}, \hat{B} \rangle$ above, we obtain in the steady state

$$\langle : \Delta\hat{P}(t), \Delta\hat{P}(t+\tau) : \rangle \overset{t\to\infty}{=} \frac{1}{2}P_e(\infty)M(\tau) + \left(\langle \Delta\hat{P}(\infty) \rangle \langle \Delta\hat{P}(\tau) \rangle - \langle \Delta\hat{P}(\infty) \rangle \langle \Delta\hat{P}(\infty) \rangle\right), \tag{11}$$

where P_e and $\Delta\hat{P}$ are obtained from Eqn. 4 by setting $t \to \infty$. It is only reasonable that in the steady-state, the two-time correlation depends only upon the delay, not upon the initial time t - i.e., the fluctuating fluorescent field exhibits "stationarity". In passing, we also note from Eqn. 11 the reason for including the "comma" in the definition of the squeezing spectrum in Eqn. 10: Subtraction of the time-independent term $\langle\Delta\hat{P}(\infty)\rangle\langle\Delta\hat{P}(\infty)\rangle$ ensures the absence of delta-function like contributions, an unwanted feature if one were examining squeezing.

Substituting Eqn. 11 in Eqn. 10, we find that the squeezing spectrum $S_\phi(\omega)$ can be expressed as a sum of two terms, $S_{f.e.}(\omega)$ and $S_{p.e.}(\omega)$, arising from spontaneous emission from the fully excited state and the partly excited state respectively:

$$\begin{aligned} S_{f.e.}(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \frac{1}{2} P_e(\infty) M(\tau) e^{i\omega\tau} \\ &= \frac{2\gamma^2\Omega^2}{2\Omega^2+\gamma^2}\left[\frac{1}{\omega^2+\gamma^2/4}\cos^2\theta + \frac{2\Omega^2+\gamma^2+\omega^2}{\left(\Omega^2+\gamma^2/2-\omega^2\right)^2+(\frac{3}{2}\gamma\omega)^2}\sin^2\theta\right], \end{aligned} \tag{12}$$

and

$$\begin{aligned} S_{p.e.}(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \left[\langle\Delta\hat{P}(\infty)\rangle\langle\Delta\hat{P}(\tau)\rangle - \langle\Delta\hat{P}(\infty)\rangle\langle\Delta\hat{P}(\infty)\rangle\right] \\ &= \frac{2\gamma\Omega^2\sin^2\theta}{2\Omega^2+\gamma^2}\left[\frac{2\gamma(\Omega^2-\gamma^2-\omega^2)}{\left(\Omega^2+\gamma^2/2-\omega^2\right)^2+(\frac{3}{2}\gamma\omega)^2}\right], \end{aligned} \tag{13}$$

where we have used
$\langle\Delta\hat{P}(\infty)\rangle\langle\Delta\hat{P}(\tau)\rangle - \langle\Delta\hat{P}(\infty)\rangle\langle\Delta\hat{P}(\infty)\rangle = -4a_1(\infty)\left\{a_1(\tau)-a_1(\infty)\right\}\sin^2\theta$ based on Eqns. 3 and 4.

We can easily see from Eqn. 12 that $S_{f.e.}(\omega)$ is always positive, hence cannot yield any squeezing. On the other hand, Eqn. 13 tells us that $S_{p.e.}(\omega)$ may go negative for certain values of Ω and γ. This negative value is maximum when $sin^2\theta$ is maximum, i.e., when $\theta = 90^0$ Adding $S_{f.e.}(\omega)$ and $S_{p.e.}(\omega)$ together we obtain the squeezing spectrum $S_\phi(\omega)$[3]:

$$S_\phi(\omega) = \frac{2\gamma^2\Omega^2\sin^2\theta}{2\Omega^2+\gamma^2}\left[\frac{1}{\omega^2+\gamma^2/4}\cos^2\theta + \frac{4\Omega^2-\gamma^2-\omega^2}{\left(\Omega^2+\gamma^2/2-\omega^2\right)^2+(\frac{3}{2}\gamma\omega)^2}\sin^2\theta\right]. \tag{14}$$

Equation 14 corrects a couple of misprints in the expression for $S_\phi(\omega)$ given in Ref.3, Equations 11.51 and 11.52. We see that maximum squeezing occurs at $\theta = 90^0$ for weak excitation $\Omega^2 < \frac{\gamma^2}{4}$, and is contributed *entirely* by $S_{p.e.}(\omega)$ (Eqn. 13).

Thus we see that it is the spontaneous emission from the partly excited atom which is responsible for causing phase-sensitive squeezing in the resonance fluorescence of weakly driven short-lived atoms on-resonance. In the following we shall see that the same process causes squeezing in strongly driven long-lived atoms as well.

4.2. The Long-lived Atom

In the absence of damping, the mean fluorescent intensity never settles into a "steady state", instead continuing to oscillate at the Rabi frequency for the entire duration of the interaction time τ_0. The two-time correlation function is of course given by Eqn. 5. To evaluate the spectrum it is necessary to average eqn. 5 over the initial photodetection time t for $0 \le t \le \tau_0 - \tau$ where τ ranges from 0 to τ_0. For strongly excited long-lived atoms $\Omega'\tau_0 \gg 1$, all terms in Eqn. 5 containing $\sin\Omega' t$ or $\cos\Omega' t$ are eliminated by the average over t (typically many Rabi oscillations occur during the interaction time). In this case, the squeezing spectrum is defined as

$$S_\phi(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \langle : \Delta\hat{P}(t), \Delta\hat{P}(t+\tau) : \rangle_t e^{i\omega\tau}, \tag{15}$$

where $\langle \ldots \rangle_t$ denotes the average over t. We note that because the radiative lifetime far exceeds the interaction time, the photon detected at time $t+\tau$ *is* the next emitted photon after the first one at t, i.e., the probability

is negligible that there was a spontaneous emission between t and $t+\tau$ which was not detected. Because the system never settles into a steady-state in the case of the long-lived atom, there is no $t \to \infty$ type long-time contribution (like the $\langle \Delta\hat{P}(\infty)\rangle\langle \Delta\hat{P}(\infty)\rangle$ term in Eqn. 13 for the case of the short-lived atom above), meaning that in the case of the long-lived atom we can dispense with the comma in Eqn. 15, and simply write

$$\begin{aligned} S_\phi(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \langle :\Delta\hat{P}(t)\Delta\hat{P}(t+\tau):\rangle_t e^{i\omega\tau} \\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \left[\frac{1}{2}\langle P_e(t)\rangle_t M(\tau) + \left(\langle \Delta\hat{P}(t)\rangle_t \langle \Delta\hat{P}(\tau)\rangle_g\right)\right] \end{aligned} \tag{16}$$

Therefore, for long-lived atoms, neglecting relaxation effects, the squeezing spectrum defined in Eqn. 15 is, for all practical purposes, identical to the frequency spectrum, i.e., the Fourier transform of the phase-sensitive autocorrelation.

Just as in the case of short-lived atoms, we find (upon substituting Eqn. 17 in Eqn. 15) that the squeezing spectrum $S_\phi(\omega)$ in Eqn. 15 can be again expressed as a sum of two terms, $S_{f.e.}(\omega)$ and $S_{p.e.}(\omega)$ arising from spontaneous emission from the fully excited state and the partly excited state respectively:

$$\begin{aligned} S_{f.e.}(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \frac{1}{2}\langle P_e(t)\rangle_t M(\tau) e^{i\omega\tau} \\ &= \frac{\Omega^2\tau_0}{8\pi\Omega'^2}\left[L(\delta\tau_0)\cos^2\phi + \left(\frac{\Delta^2}{\Omega'^2}L(\delta\tau_0) + 2\frac{\Omega^2}{\Omega'^2}L(\omega\tau_0)\right)\sin^2\phi\right], \end{aligned} \tag{17}$$

and

$$\begin{aligned} S_{p.e.}(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\tau \langle \Delta\hat{P}(t)\rangle_t \langle \Delta\hat{P}(\tau)\rangle \\ &= \frac{\Omega^2\tau_0}{8\pi\Omega'^2}\left[\frac{\Delta^2}{\Omega'^2}\sin^2\phi\left\{2L(\omega\tau_0) - L(\delta\tau_0)\right\} + \frac{\Delta}{\Omega'}\frac{\sin(\delta\tau_0) - \delta\tau_0}{(\delta\tau_0)^2}\sin 2\phi\right]. \end{aligned} \tag{18}$$

where $P_e(t)$ and $\langle \Delta\hat{P}(t)\rangle$ are given by Eqns. 8 and 6 respectively. Here $\delta \equiv \omega - \Omega'$ and $L(x) \equiv \frac{\sin^2 x}{x^2}$.

In analogy to the case of the short-lived atom, we see from Eqn. 17 that $S_{f.e.}(\omega)$ is always positive, hence cannot yield any squeezing. On the other hand, Eqn. 18 tells us that $S_{p.e.}(\omega)$ may become negative for certain values of Ω and Δ, but we have to be careful: When we add $S_{f.e.}(\omega)$ and $S_{p.e.}(\omega)$ to obtain the squeezing spectrum $S(\omega)$, we see that the "$-L(\delta\tau_0$" term in Eqn. 18 is exactly cancelled by a corresponding term in Eqn. 17, hence the squeezing does not arise from this term. In fact, the squeezing arises entirely from the $\sin 2\phi$ term. For $\Delta\sin 2\phi \geq 0$, the $\sin\delta\tau_0 - \delta\tau_0$ term goes negative. Maximum squeezing is obtained by setting $\phi = 45^0$. However, note that the $\sin 2\phi$ term, and hence the squeezing for the long-lived atom, is zero if the detuning $\Delta = 0$. The role of non-zero detuning is to suppress the likelihood of the atom being in the fully excited state, thus enhancing the contribution of the fluorescent field from a partially excited atom and hence the squeezing.

To summarize this section, we have calculated the squeezing spectrum for both the short-lived and the long-lived atom. The squeezing in the resonance fluorescence from a short-lived atom is maximized for weak, on-resonant excitation, and is detected in the quadrature that is out-of-phase[1] with the driving field. By contrast, the squeezing in the resonance fluorescence from a long-lived atom is maximized for strong, off-resonant excitation, and is detected in the 45^0 quadratures. However, despite this major difference between short- and long-lived atoms, the squeezing arises in both cases from the $S_{p.e.}(\omega)$ term, specifically the $\langle \Delta\hat{P}(\tau)\rangle_g$ term in Eqn. 5, which describes the following process: Spontaneous emission from a *partly* excited atom at t causes a quantum collapse of the atomic wavefunction to the ground state, followed by spontaneous emission of another photon at time τ later.

5. CONCLUSIONS

In conclusion, we have measured phase-sensitive temporal correlations in resonance fluorescence which help elucidate the phase properties of the fluorescent field. Two distinct contributions to the field autocorrelation

function are identified, one corresponding to atomic emission from a partly excited state, and the other from the fully excited state. Differences between the phase properties of the two terms are analyzed using simple physical arguments. Further, we have shown that writing the correlation function in the form of Eqn. 5 is important because it enables us to keep track of which physical process during phase-dependent resonance fluorescence gives rise to the phenomenon of squeezing. We find that despite major differences in the circumstances under which squeezing is observable in a long-lived and a short-lived atom, the physical process responsible for squeezing is one and the same.

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