Observation of Linewidth Narrowing Due to Coherent Stabilization of Quantum Fluctuations

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Quantum fluctuations of an optical transition moment are observed to be suppressed by strong coherent excitation of a weak auxiliary transition. The stabilization is manifest through the appearance of subnatural linewidths in the resonance fluorescence spectrum and is due to coherent mixing of atomic states. Our results are in quantitative agreement with the predictions of L. M. Narducci *et al.* [Phys. Rev. A **42**, 1630 (1990)].

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It has been shown that the quantum fluctuations exhibited by atoms contained within optical or microwave cavities can be dramatically different from those exhibited by atoms in free space.[1] Recently, Narducci *et al.*[2] predicted that certain quantum fluctuations associated with three-level atoms can be significantly modified *without* the use of a cavity by exposing the atoms to a strong coherent-state driving field.[3] This prediction is intriguing because it opens up to researchers a new class of systems in which quantum fluctuations can be manipulated and the effects directly observed. In the present experimental work, a stabilization of quantum fluctuations, manifest in a narrowing of resonance fluorescence linewidths to subnatural values, is reported. Agreement with theory is excellent.

The atomic system considered by Narducci *et al.*[2] consists of a V-configuration, three-level atom[4] [see Fig. 1(a)], with ground state $|g\rangle$ and excited states $|s\rangle$ and $|w\rangle$. The excited states, $|s\rangle$ and $|w\rangle$, decay back to the ground state at the rates γ_s and γ_w, respectively. The $|g\rangle \rightarrow |s\rangle$ ($|g\rangle \rightarrow |w\rangle$) transition is driven by a resonant, coherent-state, driving field of frequency ω_s (ω_w) and Rabi frequency Ω_s (Ω_w). It is assumed that the $|g\rangle \rightarrow |w\rangle$ transition is much weaker than the $|g\rangle \rightarrow |s\rangle$ transition (i.e., $\gamma_w \ll \gamma_s$). In Ref. 2, it is shown that the light inelastically scattered by the atom in the spectral vicinity of the strong transition frequency ω_s displays a spectral narrowing (i.e., stabilization of the quantum fluctuations) when the weak transition is driven more and more strongly. For $\Omega_w \gg \Omega_s \gg \gamma_s$, the strong-transition inelastic fluorescence spectrum consists of a central component with a spectral full width at half maximum (FWHM) of γ_w and two symmetric sidebands of width $3\gamma_w/2$. This result is surprising because the inelastic spectrum scattered by the atom for $\Omega_w = 0$ and $\Omega_s \gg \gamma_s$ (the Mollow spectrum[5]) has features with FWHM widths of γ_s (central peak) and $3\gamma_s/2$ (sidebands).

Our experiment involved measuring the resonance fluorescence spectrum emitted on the stronger of two transitions within V-configuration, three-level-like, barium atoms as depicted in Fig. 1(a). The strong (weak intercombination line) $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_1$ ($6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^3P_1$) transition has a frequency denoted by ω_{sg} (ω_{wg}), a spontaneous lifetime of 8.37 ± 0.08 nsec (Ref. 6) [3.35 ± 0.5 μsec (Ref. 7)], a resonance wavelength of 553.5 nm (791.1 nm), and is hereafter called the strong (weak) transition. The coherent-state driving fields were derived from the outputs of two actively stabilized ring dye lasers (Coherent 699-21, linewidth ≈ 1 MHz) and were collimated, superimposed, and made incident on a Ba atomic beam which passed through the center of a 50-cm confocal Fabry-Pérot interferometer. The finesse of the Fabry-Pérot interferometer was ≈ 100 based on the measured ≈ 1.5-MHz transmission linewidth. The atomic beam had a residual Doppler width of ≈ 3 MHz. In the interaction region, the atomic, weak-transition laser, and strong-transition laser beams had diameters (FWHM) of 200 μm, 750 μm, and 2 mm, respectively. The weak- and strong-transition laser beams had nearly Gaussian profiles and had maximum powers of 200 and 20 mW, respectively. The laser beams, the atomic beam, and the cavity axis were mutually orthogonal [see Fig. 1(b)].

The fluorescence spectrum of the atoms was obtained by recording the fluorescence intensity emitted out the ends of the confocal Fabry-Pérot interferometer as a function of the Fabry-Pérot mirror spacing (i.e., trans-

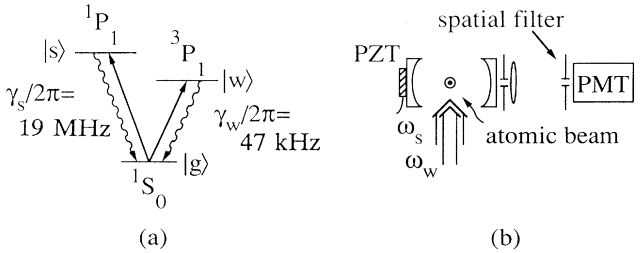

FIG. 1. (a) Energy-level structure of a ^{138}Ba atom. γ_s (γ_w) is the spontaneous decay rate of the $^1P_1$ ($^3P_1$) state. ω_{sg} (ω_{wg}) is the $^1S_0 \rightarrow {}^1P_1$ ($^1S_0 \rightarrow {}^3P_1$) transition frequency. (b) Experimental setup. The cavity is used to increase the fluorescence signal that reaches the detector and does not significantly modify the total spontaneous emission rate of the atom.

mission frequency). At the low-atomic-number densities used in the present experiment, the measured spectrum corresponds to the single-atom resonance fluorescence spectrum with contributions from both elastic and inelastic scattering.[8] Imaging and spatial-filtering techniques were employed to select only that light which was emitted nearly orthogonal to the atomic beam and which originated from a source volume of approximately 850 μm in diameter. Spectral filtering was employed to block light scattered at the weak-transition frequency. We stress that placing the atomic beam inside the confocal Fabry-Pérot cavity merely serves to facilitate measurement of the fluorescence spectrum and, in the configuration employed, does not significantly affect atomic relaxation. The instrumental resolution, which is limited by the combined effects of cavity linewidth, laser linewidth, and Doppler broadening, was equal to 8.5 ± 0.5 MHz. This value was determined from measurements of the apparent linewidth of elastically scattered resonance fluorescence[9] generated via weak excitation of the strong transition (i.e., $\Omega_w = 0$, $\Omega_s \ll \gamma_s$).

When only the strong transition of the atoms is driven, a classic Mollow-type, three-peaked resonance fluorescence spectrum is expected. This spectrum has contributions from both elastically and inelastically scattered light. The elastic contribution decreases in importance as the driving-field strength is increased, has negligible spectral width, and contributes only to the central component of the Mollow triplet. Inelastic scattering contributes to all three peaks and is responsible for the ultimate strong-field widths that were given above. In Ref. 2, it is predicted that excitation of the weak transition will modify the spectrum emitted on the strong transition by changing the fractional contributions of elastic and inelastic scattering (generally decreasing the importance of the former), and by narrowing the spectral width of features arising from inelastic scattering. Our primary objective is to measure the latter effect. Ideally, measurements involving the Mollow sidebands would reveal this effect uncomplicated by elastic scattering. Unfortunately, the widths of these peaks are strongly affected by difficult-to-avoid spatial variations in the excitation-field intensities and we therefore concentrate our analysis on the behavior of the central-peak linewidth.

Shown in Fig. 2 are measured strong-transition fluorescence spectra (dashed lines) obtained for one value of the strong-transition Rabi frequency ($\Omega_s \simeq 35$ MHz) without [Fig. 2(a)] and with [Fig. 2(b)] simultaneous excitation of the weak transition. The dot-dashed curve in Fig. 2(a) was generated using a theoretical expression for the Mollow spectrum convolved with the measured instrumental response function. The strong-transition Rabi frequency and the vertical scale factor were taken as free parameters. The solid line in Fig. 2(a) was calculated in the same manner as the dot-dashed line except that the adjacent transmission peaks

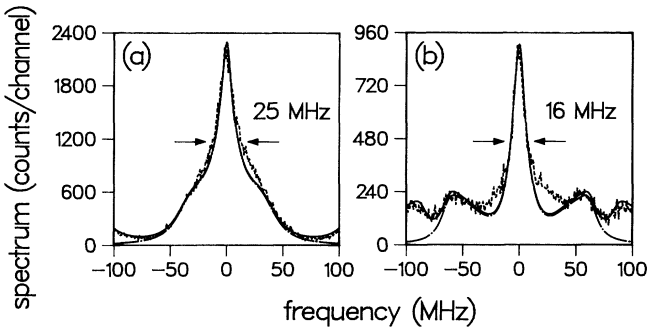

FIG. 2. Measured (dashed line) and calculated (solid and dot-dashed lines) fluorescence spectra near the $^1S_0 \rightarrow {}^1P_1$ transition frequency. (a) $\Omega_s \simeq 35$ MHz, $\Omega_w = 0$, the spectrum corresponds to the monochromatic excitation of two-level atoms. (b) $\Omega_s \simeq 35$ MHz, $\Omega_w \simeq 43$ MHz, the spectrum is significantly modified from that of (a). See the text for a discussion of the significance of the observed 16-MHz spectral width. The channel width used in the experiment was 0.8 MHz.

of the 150-MHz free-spectral-range Fabry-Pérot analyzer were included in the instrumental response function. Excellent agreement between the calculated and observed spectra is obtained when the free-space linewidth used in the theoretical expression for the Mollow spectrum is taken to be 19 MHz. This value is equal to the best independently measured values of the quantity.[6]

In Fig. 2(b), both transitions are driven ($\Omega_w \simeq 43$ MHz). It is clear that excitation of the weak transition dramatically modifies the spectrum of fluorescence emitted on the strong transition. The sideband peaks are spread further apart, and most interestingly, the measured FWHM of the central peak drops to 16 MHz. The dot-dashed curve in Fig. 2(b) was calculated following Ref. 2 generalized to take into account elastic scattering[10] using the same values for the strong-transition natural linewidth and Rabi frequency that were employed in the corresponding calculation in Fig. 2(a). The weak-transition natural linewidth employed in the calculation was deduced from the upper-state lifetime given above. Variations in the weak-transition excitation-field intensity (important because of the small diameter of that beam) were accounted for by averaging the calculated spectrum across the weak-excitation-field cross section. The resulting spectrum was convolved with the instrumental response function. In the simulations, the average value of the weak-transition Rabi frequency and the vertical scale factor were allowed to vary so as to maximize the fidelity. The best-fit value of the former was consistent with experimental measurements. In addition, the best-fit vertical scale was within 5% of that used to generate Fig. 2(a).

The data shown in Fig. 2(b) clearly demonstrate that the linewidth narrowing is a pronounced effect since the 16-MHz FWHM of the central peak in Fig. 2(b) is less than the 19-MHz free-space natural linewidth of the

strong transition even before deconvolving the instrumental response function. Our best estimate of the actual width of the inelastic contribution to the central peak is 7 MHz. The width corresponds to the width of the calculated spectrum (dot-dashed line) prior to convolution with the instrumental response function. We note that our simulations explicitly account for elastic scattering, and indicate that it cannot account for the narrowness of the central peak observed in Fig. 2(b). We thus conclude that the quantum fluctuations of the strong-transition moment are dramatically stabilized by the application of the weak-transition driving field. The solid line in Fig. 2(b) was calculated in the same manner as the solid line in Fig. 2(a).

In Fig. 3, strong-transition fluorescence spectra obtained with a higher strong-transition Rabi frequency are shown. In Fig. 3(a) ($\Omega_s \simeq 55$ MHz, $\Omega_w = 0$), we see that at this larger value of Ω_s the classic triplet structure of the Mollow spectrum begins to be apparent. In Fig. 3(b), a strong-transition spectrum obtained with excitation on both the weak and strong transitions ($\Omega_s \simeq 55$ MHz, $\Omega_w \simeq 43$ MHz) is presented. It is seen that the application of the weak-transition driving field increases the sideband spacing in the strong-transition fluorescence spectrum and causes a narrowing of the central peak. The dot-dashed and solid lines represent simulations calculated according to the procedure described in the case of Fig. 2. The simulations provide a good description of the observed spectra. From the best-fit simulations, we deduce that the widths of the inelastic components of the central peaks shown in Figs. 3(a) and 3(b) are 21 and 12 MHz, respectively. The 21-MHz width found in the former case is slightly larger than the 19-MHz natural width of the strong transition and results from the close proximity of the still-not-fully-resolved Mollow sidebands. In the case of Fig. 3(b), the 12-MHz deduced width of the central peak's inelastic component is significantly narrower than the strong-transition natural width.

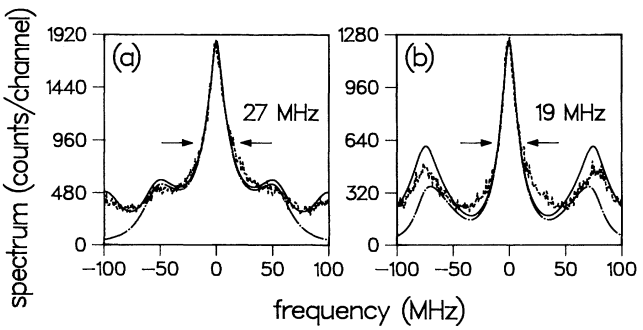

FIG. 3. Measured (dashed line) and calculated (solid and dot-dashed lines) fluorescence spectra near the $^1S_0 \rightarrow {}^1P_1$ transition frequency. (a) $\Omega_s \simeq 55$ MHz, $\Omega_w = 0$. (b) $\Omega_s \simeq 55$ MHz, $\Omega_w \simeq 43$ MHz, the spectrum is significantly modified from that of (a).

In limiting cases, the stabilization effect observed here can be calculated in a rather straightforward manner (see Ref. 2 for the general case). We first derive the semiclassical equations of motion for the expectation values of the density operator in a transformed basis. The eigenstates in the transformed basis are given in terms of the energy eigenstates of the undriven atom through the relation

$$\begin{pmatrix} |g(t)\rangle \\ |c(t)\rangle \\ |d(t)\rangle \end{pmatrix} = \frac{1}{\Omega} \begin{pmatrix} \Omega & 0 & 0 \\ 0 & \Omega_s e^{i\omega_s t} & \Omega_w e^{i\omega_w t} \\ 0 & \Omega_w e^{i\omega_s t} & -\Omega_s e^{i\omega_w t} \end{pmatrix} \begin{pmatrix} |g\rangle \\ |s\rangle \\ |w\rangle \end{pmatrix}, \quad (1)$$

where the generalized Rabi frequency is given by $\Omega = (\Omega_s^2 + \Omega_w^2)^{1/2}$. We have assumed, for simplicity, that Ω_s and Ω_w are real quantities. The equations of motion in the transformed basis take on a particularly simple form for the case of intense, resonant excitation of the strong and weak transitions ($\Omega_w \gg \Omega_s \gg \gamma_s, \gamma_w$) and are approximately given by

$$\frac{d}{dt}\sigma_{cg} = -\frac{\gamma}{2}\sigma_{cg} - i\frac{\Omega w_{cg}}{2} \quad (2a)$$

and

$$\frac{d}{dt}w_{cg} = -\gamma(w_{cg}+1) - i\Omega(\sigma_{cg} - \sigma_{gc}), \quad (2b)$$

where the slowly varying coherence and the inversion between the states are denoted by σ_{cg} and w_{cg}, respectively, and $\sigma_{gc} = \sigma_{cg}^*$. The effective, coherently modified decay rate is defined as

$$\gamma = \frac{\Omega_s^2}{\Omega^2}\gamma_s + \frac{\Omega_w^2}{\Omega^2}\gamma_w. \quad (3)$$

Physically, the state $|d(t)\rangle$ is decoupled from the other states[11] and any population initially in the state $|d(t)\rangle$ is rapidly pumped into the other states. Hence, the one set of equations (2) completely describes the interaction of the two, intense coherent fields with the three-level atom. Note that Eqs. (2) are *identical* to the equations of motion for a two-level atom characterized by a population decay rate γ, driven by a resonant, monochromatic field of Rabi frequency Ω.[12] These equations are valid to order $\Omega_s\gamma_s/\Omega_w^2$ and are consistent with the approximate results derived in Ref. 2.

The inelastic spectra for the strong and weak transitions can be derived from the transformed equations of motion (2) through use of the quantum regression theorem.[2,10] Alternatively, the spectra could be derived using the dressed-state formalism.[13] It is found that the functional dependence of the spectra for the two different transitions are the same and that only the scaling is different. As could be inferred from Eq. (2), the spectrum is that of a two-level atom, where the width of the central component of the triplet is equal to γ and the

sideband spacing is equal to Ω. Hence, the widths of the spectral features for the strong (weak) transition are narrower (broader) than they would be in comparison to the case when either of the transitions is excited individually (since $\gamma_s \geq \gamma \geq \gamma_w$). Therefore, the coherent coupling between the atomic levels suppresses the quantum fluctuations of the strong transition at the expense of increasing the fluctuations of the weak transition. We have verified that our experimentally measured spectra showing dramatic spectral narrowing are in good agreement with the approximate model just presented.

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