Vacuum-Field Dressed-State Pumping

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The steady-state behavior of two-level atoms, driven by a strong field and relaxing into a frequency-dependent vacuum reservoir, has been studied in an experiment involving Ba atoms in a confocal cavity. Enhancement or suppression of the atomic excitation is observed when the cavity is tuned to either of the strong-field fluorescence sidebands. These effects result from a redistribution of the population among the dressed atom-field states induced by the cavity-modified vacuum field. The enhancement effect may be useful in achieving significant steady-state inversion.

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The influence of environment on spontaneous radiative decay properties has attracted considerable attention in recent years. It has been predicted[1] that cavity-confined atoms may experience an inhibition of spontaneous emission because of a cavity-induced reduction in resonant electromagnetic-mode density. The veracity of this prediction has been demonstrated in a number of experiments in the microwave,[2,3] infrared,[4] and optical[5-8] regimes. The opposite effect in which the spontaneous decay rate is enhanced over its free-space value because of a cavity-induced increase in mode density has also been reported.[6,8,9] These results have stimulated a number of theoretical works related to modified spontaneous emission under various special circumstances.[10-14]

In all of the work above, with the exception of the recent radiative line-shift measurements,[6] the spectral structure of the cavity-modified electromagnetic-mode density (i.e., of the vacuum reservoir) has not played a significant role. Entirely new phenomena including dynamical suppression of spontaneous emission,[15,16] non-Lorentzian radiative line shapes,[17] dressed-state pumping,[16] and atomic squeezing[18] have been predicted to occur in cases where the vacuum reservoir is frequency dependent on a scale comparable to or finer than the atomic resonance width. These effects are especially interesting because they can be viewed as resulting from the existence of a finite memory time in the cavity-modified vacuum reservoir.

We report here the first experimental study of atomic relaxation dynamics as perturbed by a frequency-dependent vacuum reservoir. Specifically, we have studied the modification of excited-state population in strongly driven atoms induced by tuning a cavity resonance, which corresponds to a peak in the vacuum-mode density, across the atomic fluorescence line shape. It is found that atomic excitation can be enhanced or suppressed depending on the spectral location of the vacuum-mode-density peak relative to the center of the atomic fluorescence line.

A simple theoretical description of the basic phenomena involved can be obtained by consideration of a two-level atom having a transition frequency ν_0 and a ground (excited) state $|g\rangle$ ($|e\rangle$) with population n_g (n_e) that is driven by a strong optical field of frequency ν_l. Let $\Delta = (\nu_l - \nu_0)$. The atom-field system is assumed to decay via coupling to a frequency-dependent vacuum reservoir. We describe our system in terms of the atom-field dressed states,[19,20] which consist of a ladder of doublets separated in frequency by ν_l and split by the generalized Rabi frequency (see Fig. 1). In the dressed-atom picture, the spontaneous-emission process consists of transitions between levels of adjacent doublets and the creation of reservoir photons. At steady state, we must have[19]

$$\Gamma_{12}\Pi_1 = \Gamma_{21}\Pi_2 , \qquad (1)$$

where Π_1 (Π_2) represents the total population, summed over the photon number, of the upper (lower) component of the dressed-state doublet and Γ_{12} and Γ_{21} are spon-

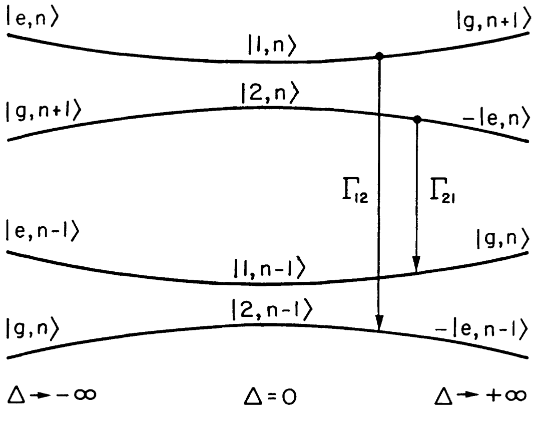

FIG. 1. Schematic energy diagram of two atom-field dressed-state doublets. The product states at the right (left) represent asymptotic forms at large positive (negative) Δ.

taneous decay rates (see Fig. 1). We may write the decay rates in the form

$$\Gamma_{ij} = \rho_{ij} d_{ij} \quad (i,j = 1,2) ; \tag{2}$$

here d_{ij} is proportional to the square of the atomic dipole matrix element between the considered dressed states and ρ_{ij} is a factor proportional to the photon-mode density at the corresponding transition frequency. After evaluation of the atomic dipole matrix elements, one can immediately deduce the steady-state values of Π_1 and Π_2, and, consequently, of n_e:

$$n_e = \frac{\rho_{21} \cos^2\theta \sin^4\theta + \rho_{12} \cos^4\theta \sin^2\theta}{\rho_{21} \sin^4\theta + \rho_{12} \cos^4\theta} , \tag{3}$$

where $\tan(2\theta) = -\Omega_R/\Delta$ $(0 \leq \theta \leq \pi/2)$ and Ω_R is the on-resonance Rabi frequency. Equation (3) states that we can control the excited-state atomic population by changing the relative values of ρ_{12} and ρ_{21}. The imbalance between the mode densities at the frequencies corresponding to the 12 and 21 transitions results in an optical pumping of the dressed state and consequently of the bare atomic states. This effect, which one might term *vacuum-field dressed-state pumping*, was predicted by Lewenstein and Mossberg.[16] Significantly, note that for

$$\rho_{12}/\rho_{21} < \tan^4\theta \quad (\Delta < 0) ,$$

or

$$\rho_{12}/\rho_{21} > \tan^4\theta \quad (\Delta > 0) ,$$

a steady-state positive atomic inversion can be realized. If $\rho_{ij}/\rho_{ji} = 10$, a maximum steady-state inversion $(n_e - n_g)$ of $\simeq 0.15$ is predicted. In our experiment, ρ_{ij} and ρ_{ji} differed by only 5% and no attempt was made to ascertain whether or not the small positive inversion expected did in fact occur.

In our experiment, a beam of atomic Ba was made to pass through the center and normal to the axis of a 1-cm symmetric confocal optical cavity. The mode degeneracy of this type of cavity is known[5] to give rise to a frequency-dependent photon-mode density. The atomic beam was 200 μm in diameter and had a collimation of 1:280. The cavity mirrors were spherical with 1-cm radius of curvature and had a specified reflectivity of 99.3%. Cavity finesse measured with a collimated 1-mm-diam laser beam was typically 180. The cavity mirrors were mounted in an Invar holder which was equipped with a piezoelectric transducer for cavity tuning and temperature stabilized to 10 mK. The reflecting surface of one of the 5-mm-diam mirrors was limited by an intracavity aperture of 2.2 mm in diameter. The 553.5-nm $6s^2\,{}^1S_0$-$6s6p\,{}^1P_1$ barium resonance line was driven by the output of a cw ring dye laser propagating normal to both the cavity axis and the barium beam. With a saturation resonance in an auxiliary Ba vapor cell, the laser frequency could be actively locked anywhere within ±500 MHz of ν_0. Laser-field inhomogeneity was minimized by our passing the beam through an aperture and then imaging the aperture into the laser-atom interaction region. In the interaction region, the laser beam had a 150 μm diameter and $\simeq 10$ mw power. At any instant, approximately 50 atoms were within the interaction volume. The magnetic field at the interaction region was minimized with the help of external magnetic coils so that the residual Zeeman splitting was smaller than $\frac{1}{10}$ of the $\simeq 19$-MHz natural linewidth. The excitation laser was linearly polarized perpendicular to the cavity axis.

Atomic fluorescence was simultaneously monitored along the cavity axis and nearly perpendicular to it. In the latter case, imaging optics and spatial filtering were employed to collect only those photons that were emitted by atoms in a 130-μm-diam spot within the interaction region and which propagated along a direction 10° away from antiparallel with the laser beam and normal to the atomic beam. Light detected out the cavity's side was not frequency selected and thus provided a direct measure of the excited-state population of the atoms.[19] The total solid angle collected to the side was 2.5×10^{-3} sr. The two signal directions described above are referred to as the axial and side fluorescence signals, respectively.

Figure 2 shows the variation in the side fluorescence signal, generated by weak resonant excitation, as the spacing between the cavity mirrors (and hence the cavity resonance frequency) is swept. The drop in side fluorescence intensity results from a reduction in the steady-state, excited-state, and atomic population, which in turn results from a cavity-induced enhancement in the atomic spontaneous decay rate. The change in the atomic decay rate occurs because the intracavity mode density is frequency dependent (being maximum near the cavity resonance frequency) and tunes with the cavity. Importantly for the analysis that follows, in the weak excitation limit, the side fluorescence intensity is proportional to the inverse square of the mode density.[6]

Quantitative calculations concerning vacuum-field dressed-state pumping in our cavity require complete knowledge of the cavity's spatial and spectral mode distribution. Rather than attempt to calculate this distribution, we directly deduce, using the inverse square depen-

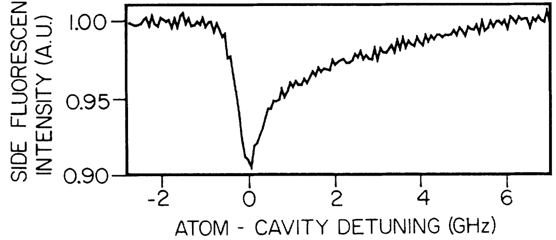

FIG. 2. Side fluorescence intensity vs atom-cavity detuning in the case of weak resonant excitation.

dence mentioned above, the spatially averaged spectral mode distribution from the cavity-modified fluorescence data of Fig. 2. Interestingly, the 10% dip in side fluorescence intensity indicates only a 5% variation in cavity-mode density as a function of frequency. The asymmetric structure of the cavity-mode distribution deduced from Fig. 2 can be qualitatively understood in terms of the geometric properties of the spherical mirror resonator.[21] Note that in the case of a weak excitation field, our vacuum reservoir is essentially constant over the atomic resonance line. This is not true in the strong-excitation-field regime reported below.

In Figs. 3(a)-3(c) (heavy solid lines), we show the measured side fluorescence intensity as a function of cavity tuning in the case of excitation by a strong laser field ($\Omega_R \simeq 600$ MHz) and with $\Delta = 320$, 0, and -320 MHz, respectively. Figure 3(d) shows the axial fluorescence signal recorded simultaneously with the side fluorescence signal of Fig. 3(a). The familiar Mollow resonance fluorescence triplet is observed.[19,20,22] The behavior of the side fluorescence in Fig. 3(b) is strikingly different from that observed in Fig. 2 in that the dip observed in Fig. 2 is completely absent. Furthermore, in Figs. 3(a) and 3(c), we see not only reductions in the side fluorescence intensity, but also enhancements. The presence of both reductions and enhancements of side fluorescence intensity indicate cavity-mediated increases in excited-state atomic population as well as decreases. It is clear that the modifications of the excited-state atomic population occur when the cavity is tuned to resonance with the sidebands of the Mollow triplet [and hence enhances the mode density at the 12 or 21 dressed-state transition frequency (see Fig. 1)]. For $\Delta > 0$ ($\Delta < 0$), *enhanced* atomic excitation occurs when the cavity is tuned to increase the mode density at the higher-frequency (lower-frequency) sideband of the resonance fluorescence triplet.

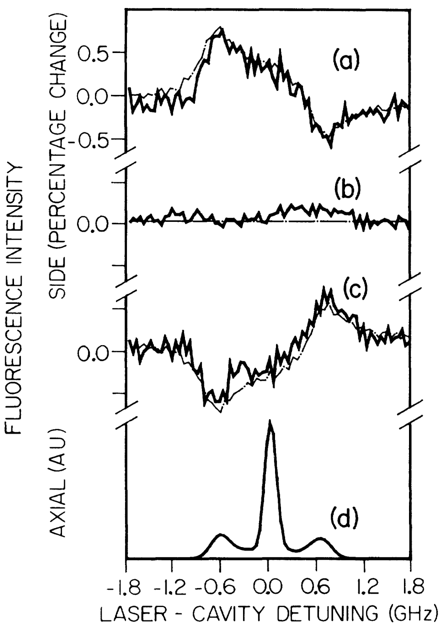

FIG. 3. (a)-(c) Side and (d) axial fluorescence intensity vs laser-cavity detuning in the case of strong excitation. Laser-atom detunings, Δ, are fixed and equal to $+320$, 0, -320, and $+320$ MHz in (a)-(d), respectively. The vertical axis for parts (a)-(c) represents percentage change relative to the average side fluorescence intensity observed for large laser-cavity detunings. In each case (a)-(c), one division represents 0.5% change. (a) and (d) were recorded simultaneously.

Interestingly, our model predicts that dressed-state pumping occurs for all three detunings studied in Fig. 3. Because of the detuning-dependent decomposition of the dressed states in terms of bare atomic states, however, at $\Delta = 0$ the expected dressed-state pumping does not lead to a change in atomic excited-state population. Note, however, that in an entirely different regime, a two-level atom interacting with a single resonant cavity mode has been predicted[23] to display a positive inversion provided that the mode is occupied by only a few photons.

To test the hypothesis that vacuum-field dressed-state pumping is responsible for the effects shown in Figs. 3(a)-3(c), we have employed Eq. (3) together with the spatially averaged spectral mode density as deduced from Fig. 2 to predict the magnitude and shape of the pumping resonance. The results of the simulation are shown as thin dash-dotted lines in Figs. 3(a)-3(c). The fluctuations in the dash-dotted line result from the use of the unsmoothed data from Fig. 2 to deduce the spectral mode density. The simulation is in excellent agreement with the observed results both in terms of magnitude and shape.

In summary, we have provided the first experimental demonstration of vacuum-field, dressed-state pumping, an effect whose existence depends on the modification of free-space spontaneous decay by a *frequency-dependent*, finite-correlation-time vacuum reservoir. By substituting highly corrected large-diameter optical components for the spherical mirrors employed in our cavity, one could reasonably expect to increase the cavity-induced change in mode density by 1 to 2 orders of magnitude compared to that observed here.[24] In this case, vacuum-field dressed-state pumping may provide for the establishment of a significant steady-state positive inversion in samples of two-level atoms.

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