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Driving the driven atom: Spectral signatures

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We have measured the emission spectrum of two-level-like Ba atoms driven by a continuous-wave bichromatic field containing a strong resonant component and a weaker component detuned from atomic resonance by the strong-field Rabi frequency. With the specified detuning, the weak field resonantly drives a transition of the atom–strong-field dressed states. Observed spectra show that each peak of the normal (single-driving-field) resonance fluorescence triplet is split into three subpeaks separated by one-half the weak-field Rabi frequency. Also seen is another triplet of peaks displaced from the atomic resonance by twice the strong-field Rabi frequency. Splitting of the normal triplet peaks can be explained through weak-field dressing of the strong-field dressed states. The origin of the additional triplet is less transparent. Comparison with theory is made. [S1050-2947(97)51411-9]

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I. INTRODUCTION

Theoretical and experimental studies of the spectral and dynamical features of two-level atoms (TLA's) under a variety of continuous-wave (cw) strong-field driving conditions have provided fundamental insight into light-matter interactions. A significant triumph of quantum optics is the prediction [1] and observation [2] of the three-peaked fluorescence spectrum of TLA's driven by a strong near-resonant monochromatic field. Of the three peaks, one occurs at the driving field frequency (the center peak), while the other two (the sideband peaks) are symmetrically displaced about the center peak by the generalized Rabi frequency. The dressed-atom model [3] emerged as a powerful description of the composite system of atom-plus-field. The system eigenstates (dressed states) form a ladder of doublets, with adjacent doublets separated by the driving field frequency and split by the generalized Rabi frequency. In this dressed-atom picture, the fluorescence peaks correspond to the transition frequencies between dressed levels, while peak areas reflect dressed level populations and relative transition strengths.

TLA's display additional absorptive and emissive spectral features and dynamics when exposed to complex or polychromatic driving fields [4]. These features have been studied in a surprisingly limited number of experiments. Application of a weak, tunable probe field to monochromatically driven TLA's demonstrated gain without population inversion [5] and observations of cw two-photon optical gain and lasing [6]. Spectrally integrated fluorescence intensity measurements [7] in the presence of driving field modulation (bichromatic excitation) showed the presence of parametric resonances. Other bichromatic field studies [8] revealed novel features such as Rabi subharmonic resonances in absorption spectra. Emission spectra dramatically different from the familiar triplet were observed in experiments involving TLA's driven by two equal intensity fields, symmetrically detuned from the atomic resonance (symmetric bichromatic excitation) [9,10]. Observed spectra display additional peaks, intensity-independent peak separation, intensity-dependent peak quantity, and alternating peak linewidths. There have also been observations of Autler-Townes spectra [11] of TLA's driven by a 100% amplitude modulated field.

Recently, Wu *et al.* [12] have experimentally investigated the transient dynamics of TLA's driven by bichromatic fields comprised of a strong and weak component. It was found that the weaker field, appropriately tuned, excites transient responses in the atom + strong-field "molecule" that are entirely analogous to those observed in the transient monochromatic excitation of purely material systems. We report here the results of a complementary experimental study of spectrally resolved fluorescence from TLA's under similar but cw driving conditions. Specifically, we make one field component resonant with the TLA (resonance frequency ν_a) and one component detuned (see Fig. 1). The detuning of the nonresonant component is equal in magnitude to the Rabi frequency of the resonant driving component. Observations reveal that many but not all aspects of the emitted spectrum

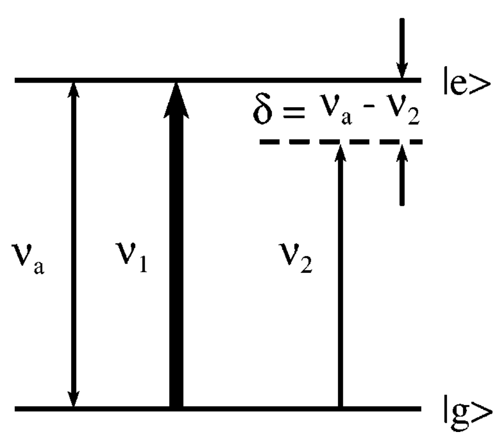

FIG. 1. The system under investigation is a two-level atom driven by a bichromatic field with one resonant, ν_1, and one off-resonant, ν_2, component.

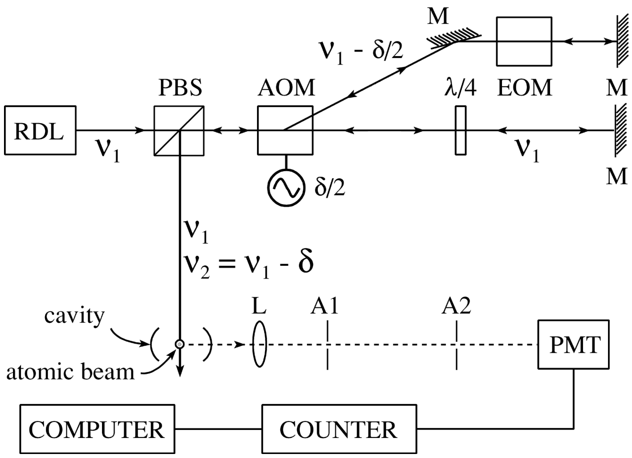

FIG. 2. Experiment schematic: RDL, cw ring dye laser; PBS, polarization beam splitter; AOM, acousto-optic modulator; M, mirror; λ/4, quarter wave plate; EOM, electro-optic modulator; L, lens; A1 and A2, apertures; PMT, photomultiplier tube.

can be understood in terms of a dressing of the dressed atom by the weaker driving field component. Recent detailed calculations of the spectra expected under our excitation conditions are in excellent agreement with our observations.

II. EXPERIMENTAL APPARATUS

Experiments were conducted in an atomic beam of natural barium with excitation frequencies chosen to maximize the contribution of nuclear-spin-free ^{138}Ba (72% abundance) to observed signals [13]. As will be discussed below, however, observed elastic-scattering signals contain substantial contributions from other isotopes. The linearly polarized output of a single-mode cw laser at frequency ν_1 (≈ 1 MHz linewidth) is frequency locked via saturation spectroscopic techniques to ν_a, the 553.5-nm $(6s^2)\ {}^1S_0 - (6s6p)\ {}^1P_1$ resonance frequency of ^{138}Ba. The laser output is double passed through an acousto-optic deflector driven at frequency $\delta/2 = 110$ MHz as shown in Fig. 2 to create a second variable amplitude driving field component at frequency $\nu_2 = \nu_1 - \delta$. With a single weak excitation field, the ^{138}Ba transition displayed an absorption linewidth of 21 MHz, while the natural width of the transition $\Gamma = 19$ MHz. Excess width is attributed to the angular spread of the atomic beam and excitation laser linewidth. It is estimated that the saturation locking scheme maintained $|\nu_1 - \nu_a| < 3$ MHz where nonorthogonality of the excitation and atomic beams contributes the primary detuning uncertainty.

The barium atomic beam (0.75 mm diameter) intersects the collimated, copropagating light fields (3.0 mm diameter) at the center of a confocal, 5-cm, Fabry-Pérot cavity. Spatial inhomogeneity of the laser excitation fields and hence Rabi frequencies over the active experimental volume is estimated to be less than 10%. The piezoelectrically scannable cavity has a finesse of 500 and a free spectral range $(c/4L)$ of 1500 MHz. The cavity axis, laser fields, and atomic beam are all aligned to be mutually orthogonal. Spectra are measured by monitoring the light power emitted out the end of the cavity as a function of cavity length. Only a single-cavity mode, whose frequency we denote by ν_c, fell within the atomic emission profile. The spectral swept rate of the cavity mode,

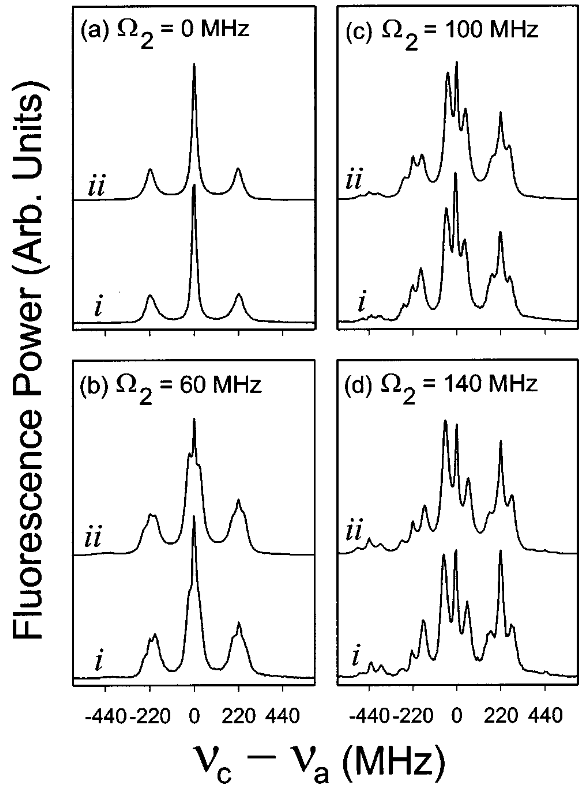

FIG. 3. Traces i (ii) represent measured (simulated) results of atomic fluorescence intensity versus cavity detuning from the atomic transition frequency $(\nu_c - \nu_a)$. $\nu_1 = \nu_a$, $\Omega_1 = \delta = 220$ MHz, and Ω_2 is specified in each plot (a)–(d).

$d\nu_c/dt$, varied by less than 3%, introducing corresponding nonlinearity in observed spectra. Experimental spectral resolution Γ_c is deduced from the observed width of weak signal elastic scattering and is found to be 13 MHz. Maximal spectral resolution is obtained through spatial and angular filtering of cavity output signal prior to photomultiplier tube detection.

III. EXPERIMENTAL MEASUREMENT

We report here (see Fig. 3) emission spectra recorded for $\Omega_1 \approx \delta = 220$ MHz and for various Ω_2, where Ω_1 (Ω_2) is the resonant Rabi frequency of the resonant (detuned) excitation field. The traces labeled i represent the measured experimental results. Figure 3(a) is the Mollow triplet produced by the strong field (ν_1) alone. When the second field is applied, the three peaks of the spectrum broaden [Fig. 3(b)] and separate into three subpeaks [Fig. 3(c)]. Each of the original peaks from the Mollow spectrum gives rise to a daughter triplet of peaks. Interestingly, the daughter triplets are split by $\Omega_2/2$ rather than Ω_2. (The value of Ω_2 is measured by temporarily setting $\nu_2 = \nu_a$, $\Omega_1 = 0$ and measuring the Mollow splitting.) As the second field intensity is further increased [Fig. 3(d)], the daughter triplets increase in splitting and an entirely new triplet of peaks appears on the far left-hand (lower frequency) side of the spectrum. There is also some evidence for an additional peak or peaks appearing on the high-frequency side of the spectrum.

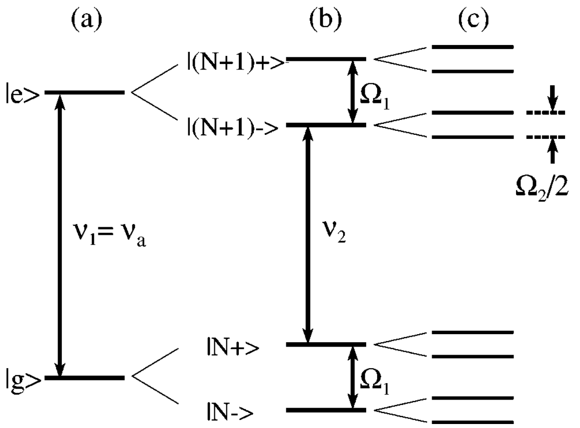

FIG. 4. (a) Undressed two-level atom; (b) ‘‘singly’’ dressed states; (c) doubly-dressed states.

IV. INTERPRETATION OF DATA

Some of the spectral features shown in Figs. 3(a)–3(d) can be derived from a simple model involving the dressing of the standard dressed states by the weaker detuned field. In Fig. 4(a), we show the levels of an undressed two-level atom. In Fig. 4(b), two doublets out of the ladder of ‘‘singly’’ dressed states ($|N\pm\rangle$) corresponding to the TLA + resonant excitation field (ν_1) are shown. The familiar resonance fluorescence triplet derives from the four possible transitions between the singly dressed states of Fig. 4(b). As indicated, the frequency of the second field, ν_2, is experimentally resonant with a transition between the singly dressed states $|N+\rangle - |(N+1)-\rangle$. One expects a second dressing of the $|N\pm\rangle$ states analogous to that introduced into the atomic states by the first field (ν_1). Resultant doubly dressed levels are shown in Fig. 4(c). Owing to the periodic structure of the singly dressed states, excitation of one singly dressed transition has the effect of splitting all of the singly dressed states giving rise to a ladder of doubly dressed quartets. When one considers all possible transitions between the quartets, one finds three triplets of peaks. The nine spectrally distinct peaks expected provide motivation for three (the center and first adjacent) triplets that appear in the experimental spectra of Figs. 3(a)–3(d). The $\Omega_2/2$ splitting of the daughter triplets is immediately motivated by calculation of the matrix element between singly dressed states and finding it to be one-half of that between the bare atomic states. The fourth triplet of peaks cannot be explained by the simple doubly dressed states of Fig. 4(c).

Recently Ficek and Freedhoff [14] have performed an extensive analysis of bichromatically driven TLA’s. Application of this theory to our specific experimental conditions predicts the spectra shown by traces *ii* in Fig. 3, where the following calculational parameters were employed: $\Gamma = 19$ MHz, $\Gamma_c = 13$ MHz, $\Omega_1 = 220$ MHz, and $\delta = 220$ MHz. Moreover, we include the effects of other weakly abundant Ba isotopes that are nonresonantly driven by the excitation fields. This consideration is important to accurately predict the peak height at ν_1 and ν_2 due to Rayleigh scattering. We point out that the theoretical plots (inclusive of isotopes) agree remarkably well with the experimental results.

If the simulation is not extended to include the multiple isotope species and the finite instrumental resolution, the spectra represented in Fig. 5 (traces *iii*) are obtained. For $\Omega_2 \neq 0$, one finds narrow peaks at $\nu_c = \nu_1 \pm n\Omega_1$, where n is an integer. These peaks represent the coherent part of spectra. We note that $\Gamma_c = 2$ MHz is assumed in this simulation to avoid divergence of the coherent peaks. It is interesting to note that the theory predicts the spectral peak at ν_a to be suppressed by the application of the field at frequency ν_2. The traces *iv* are the simulated spectra when $\Gamma_c = 13$ MHz is assumed. Instrumental resolution effectively eliminates the small-area, narrow coherent peaks. Finally, the traces *ii* (identical to those in Fig. 3) show inclusion of isotope effects as well as instrumental resolution. The primary effect of the nonresonant isotopes is to regenerate the spectral peak at frequency ν_a that is predicted to be missing in the ideal two-level atom case.

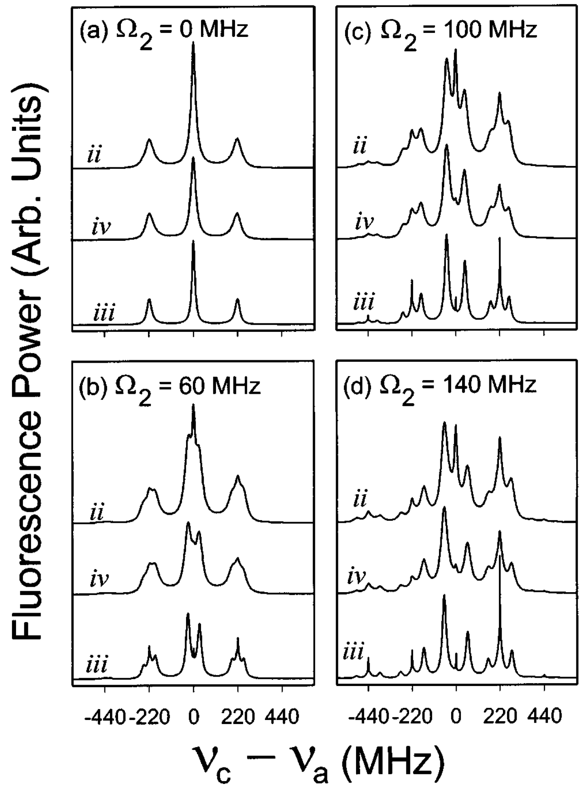

FIG. 5. Traces *iii* (*iv*) represent simulated results for $\Gamma_c = 2$ MHz (13 MHz) when only a single isotope species is considered. Traces *ii* are the same as in Fig. 3.

In conclusion, we note that TLA response to bichromatic excitation is substantially different and more subtle than it is to monochromatic excitation. The convergence of theory and experiment demonstrated here shows that this response, at least under the conditions studied here, is now reasonably well understood.

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