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Resonance fluorescence of two-level atoms under strong bichromatic excitation

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We have measured the emission spectrum of two-level-like Ba atoms driven by two, strong, equal-amplitude fields with frequency separation 2δ. The spectrum consists of a series of peaks with an essentially constant spacing δ and alternating linewidths. These features differ qualitatively from the characteristic triplet spectrum observed in the case of strong monochromatic excitation. Certain features of the observed spectrum such as its comblike structure can be motivated in terms of the energy spectrum of atom–bichromatic-field product states. Other features, such as the alternating linewidths, require more subtle analysis.

The fluorescence spectrum of two-level atoms (TLA's) driven by strong monochromatic excitation has been the subject of considerable theoretical and experimental study over the years.[1,2] This spectrum, initially predicted by Mollow[3] and subsequently observed by various groups,[4–6] possesses a characteristic symmetric triplet structure that can be inferred from the dressed-atom model.[7] The detailed understanding of the spectrum is an important achievement of quantum optics. An obvious and intriguing generalization of the Mollow problem is to determine the nature of the atomic fluorescence spectrum under conditions of bichromatic or multichromatic excitation. TLA dynamics in the presence of multichromatic excitation has been explored from a number of contexts,[8–22] but the question of resonance fluorescence has received only limited attention.[19–22] In fact, to our knowledge, no experimental work in this area has yet appeared.

We report here an experimental study of the resonance fluorescence spectrum characteristic of two-level atoms driven by a strong bichromatic excitation field. Specifically, we have studied the fluorescence spectrum of two-level-like barium atoms driven by two equal-amplitude fields that have frequencies $\nu_a-\delta$ and $\nu_a+\delta$ and are tuned near the ^{138}Ba 553.5-nm $^1S_0\rightarrow{}^1P_1$ transition frequency, ν_0. The experimental apparatus is depicted in Fig. 1. Light from a frequency-stabilized ring dye laser (Coherent 699-21) is passed through an acousto-optic modulator (AOM). The deflected and undeflected portions of the AOM output, differing in frequency by the AOM drive frequency 2δ provide the two components of the bichromatic excitation field. On emerging from the AOM, the two beams were collimated, spatially superimposed, intensity matched, and angularly aligned to maximize the depth of the 2δ intensity beat signal observed on a large-area photodiode. An 80% modulation depth was obtained. The combined, linearly polarized, beams intersected an atomic beam of natural Ba at a position coinciding with the center of a 5-cm-long confocal Fabry-Perot cavity. The Fabry-Perot is piezoelectrically scannable, has a free-spectral range of 1.5 GHz, and has an empty-cavity finesse of approximately 200. The bichromatic laser beam, Ba atomic beam, and cavity axis were mutually orthogonal. Fluorescent light transmitted out one end of the cavity was spatially filtered and imaged onto the cathode of a photomultiplier tube (PMT). The PMT output was sent to computer-controlled photon-counting electronics. In the limit of low atomic-beam densities where gain effects[23,24] can be ignored, recordings of the Fabry-Perot output intensity as a function of ν_c, where ν_c is the cavity resonance frequency, correspond to the single-atom resonance fluorescence spectrum. In our experiments, Doppler broadening and finite Fabry-Perot finesse contribute to an overall instrumental resolution of 25 MHz. The natural width of the $^1S_0\rightarrow{}^1P_1$ Ba transition is $\simeq$19 MHz.[25]

Figure 2 shows the spectra recorded for $\Delta\equiv\nu_a-\nu_0=0$, $2\delta=200$ MHz, and for various values of Ω, where Ω is the single-excitation-field-component resonant Rabi frequency. The observed spectra consist of a series of peaks symmetrically located about a central peak coinciding with the atomic transition frequency ν_0. From Fig. 2, one sees that neighboring peaks are separated in frequency by δ, and that the number of peaks increases with increasing Ω. Interestingly, the peak spacing is independent of Ω. At very low values of Ω (not shown in Fig. 2), the spectrum is observed to consist of only two peaks which correspond to elastic scattering at the excitation frequencies. These observations are consistent with the numerical cal-

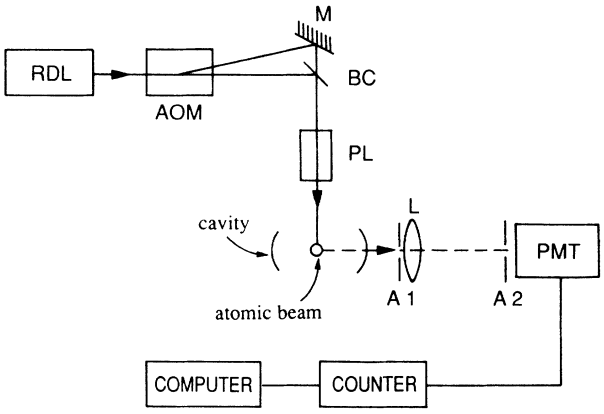

FIG. 1. Schematic of the experimental apparatus: RDL, cw ring dye laser; BC, beam combiner; AOM, acousto-optic modulator; M, mirror; PL, linear polarizer; A1 and A2, apertures; L, lens; PMT, photomultiplier tube.

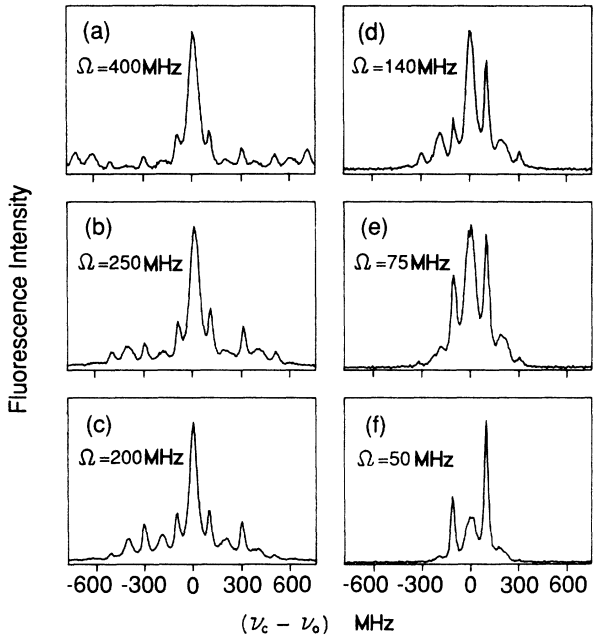

FIG. 2. Atomic fluorescence intensity vs cavity detuning from the atomic transition frequency $(\nu_c - \nu_0)$. Ba atoms are driven by a bichromatic excitation field whose components differ in frequency by 200 MHz and are located symmetrically about the atomic resonance frequency ν_0. The resonant Rabi frequency of each component of the driving field considered separately, Ω, is (a) 400 MHz; (b) 250 MHz; (c) 200 MHz; (d) 140 MHz; (e) 75 MHz; (f) 50 MHz. The small asymmetry in the spectra about ν_0 is attributed to the presence of Ba isotopes other than ^{138}Ba. Note that in (a) the 1.5-GHz free-spectral range of the Fabry-Perot cavity employed leads to distortions in the peak amplitudes observed in the portions of the spectrum most removed from ν_0.

culations of Newbold and Salamo.[20] Another interesting feature of these spectra is the alternation in linewidth as one moves from peak to peak in the spectrum. Peaks with frequencies given by $\nu_0 \pm m\delta$ with m even (m odd) have a relatively larger (smaller) linewidth. Newbold and Salamo predicted that the elastic scattering occurs only at frequencies $\nu_0 \pm m\delta$ (m odd), and for small Ω, the contributions from elastic scattering at these frequencies are larger than the contributions from inelastic scattering. Since the linewidth of the elastic scattering should be relatively narrow, its contribution to the odd-m peaks may explain the linewidth variation observed in our experiment. The observation that the number of peaks in the spectra is Rabi frequency dependent, that the peak spacings are Rabi frequency independent, and that the peak linewidths alternate all constitute features qualitatively different from those found in the case of monochromatic excitation.

A motivation for the basic structure of the observed spectra can be found in the level structure of the product states representing the uncoupled atom–bichromatic-field system. These product states can be written as $|n_a\rangle|n_1\rangle|n_2\rangle$, where $n_a = 0$ (1) represents the atomic ground state (excited state), and n_1 and n_2 represent the photon occupation numbers of the field modes at frequency $\nu_a - \delta$ and $\nu_a + \delta$, respectively. It is straightforward to show that these product states group into manifolds each corresponding to a particular value of $N = n_a + n_1 + n_2$. The centers of the manifolds are split by $h\nu_0$ (with $\nu_a = \nu_0$). Within each manifold, individual states are separated in energy by $h\delta$ (see Fig. 3). In the limit of weak excitation ($\Omega \ll \delta$) the product states should provide a good description of the system, and peaks in the fluorescence spectrum must then correspond to transitions from individual states in one manifold to states in the next lower manifold. It is clear that the only possible transition frequencies are $\nu_a \pm m\delta$, where m is an integer. Inclusion of the atom-field coupling leads to dressed states,[26] which are linear combinations of the uncoupled atom-field product states. For $\Omega \geq \delta$ one must work with the actual dressed states to predict the spectra. Interestingly, the peak positions in the observed spectra imply that the level structure of the dressed states is essentially identical to that of the product states. On the other hand, the change in the number and the relative intensity of the peaks indicates that the composition of the dressed states (in terms of the product states) depends sensitively on the atom-field interaction strength. It is interesting that the atom-field interaction modifies the composition of the dressed states without significantly perturbing their energies. While the simple energy-level considerations mentioned here provide some motivation for the locations of the peaks shown in Fig. 2, they provide no reliable insight into the alternation of peak linewidths.

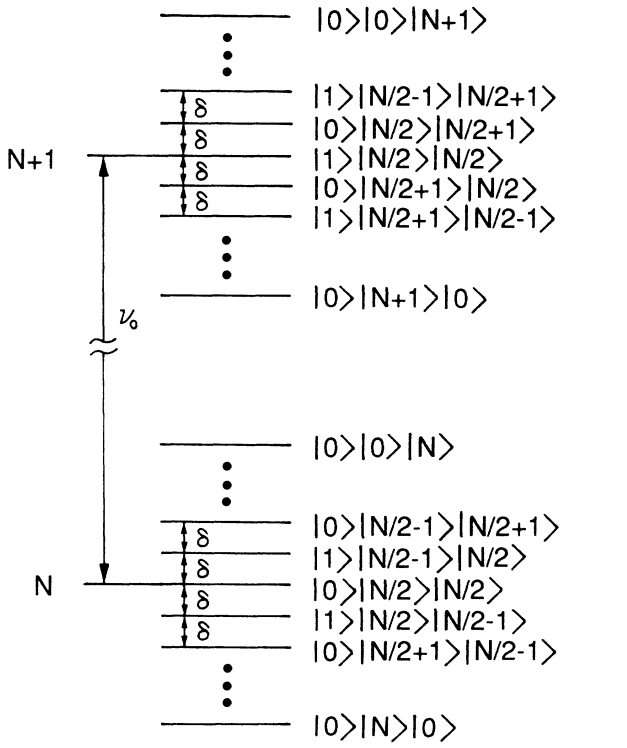

FIG. 3. Atom–bichromatic-field product states. The number $(N, N+1)$ to the left of each manifold represents the total number of excitations, i.e., $n_a + n_1 + n_2$. In the case of the lower manifold, N has been arbitrarily assumed to be even.

In conclusion, we have measured the resonance fluorescence spectrum of two-level atoms under strong bichromatic excitation. The observed spectra are qualitatively different from the Mollow spectrum. Further work involving a broader range of experimental parameters and careful comparison with the theoretical calculations is planned, and in view of the recent calculations of Wilkens and Rzazewski[22] indicating rich spectra associated with doubly amplitude modulated excitation fields, measurements are planned in this area as well.

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