Transient Spectra of Strong-Field Resonance Fluorescence

J. E. Golub
Department of Physics, Harvard University, Cambridge, Massachusetts 02138

and

T. W. Mossberg
Department of Physics, University of Oregon, Eugene, Oregon 97403
(Received 30 July 1987)

Working with a beam of two-level–atom–like ^{174}Yb, we have measured the temporal evolution of strong-field resonance fluorescence spectra during square-pulse excitation. In the case of resonant excitation, it is found that driving-field phase shifts transform the normal three-peaked Mollow spectrum into a transient doublet spectrum asymmetric with respect to the atomic transition frequency. This qualitatively new type of spectrum provides fundamental insight into the connection between atomic dynamics and spectra, and results from a nearly perfect polarization of the dressed atom-field states.

PACS numbers: 32.80.–t, 32.70–n, 42.50.Hz, 42.50.Md

The spectrum of fluorescence emitted by a two-level atom in the presence of a strong, resonant light field provides insight into basic aspects of the light-atom interaction. Because of the importance of this problem, much theoretical[1-12] and experimental[13-15] work has been addressed to it, and in the case of steady-state spectra a detailed understanding has emerged. In the case of transient spectra, the situation is quite different since theoretical predictions[16-20] have not heretofore been subjected to experimental verification. Experimental difficulties associated with the observation of transient resonance fluorescence spectra have preserved this situation despite the fact that transient spectra have been predicted to display a phenomenological richness not found in the steady state.

Early theoretical[18-20] work predicted that transient spectra would display temporal oscillations, but otherwise retain the same general three-peaked form as originally discussed by Mollow.[4] More recently, however, Lu *et al.*[16] predicted that transient spectra emitted in the course of certain resonant excitation schemes would display a qualitatively different structure. For example, when atoms are prepared in a single, well-defined, dressed state by a phase-controlled laser pulse,[21] the fluorescence spectrum is predicted to consist of two peaks instead of the three. In effect, the fluorescence intensity associated with one of the Mollow sidebands is transferred to the other, and one is left with a spectrum displaying two equal-area peaks. In this Letter, we report measurements of transient resonance fluorescence spectra obtained under a variety of experimental conditions.

As detailed by Golub, Bai, and Mossberg,[22] the transient excitation fields employed in these experiments are generated with a 200-MHz acousto-optic modulator to gate the output of a single-mode, cw, ring dye laser. The modulator, driven by a home-built rf driver, can independently switch either the amplitude or the phase of the optical excitation field. Transition times of 10 nsec are achieved. Excitation of the 556-nm $6s^2\,{}^1S_0$-$6s6p\,{}^3P_1$ transition of ^{174}Yb was carried out in a collimated atomic beam, and the radiative lifetime (875 nsec) of the upper atomic state set the time scale of the experiment. During measurements, the ring laser was frequency locked (drift <0.5 MHz/h) to a saturation resonance of an auxiliary Yb cell. By employing a large laser beam ($\simeq 5$ mm), a small atomic beam ($\simeq 0.75$ mm), and a limited fluorescence collection angle ($\simeq 10^{-5}$ sr), we ensure that the laser field is uniform throughout the sample. Fluorescence was spectrally resolved with a piezoelectrically tunable, 50-cm, confocal Fabry-Perot interferometer.[23] Transmitted photons were detected by a cooled photomultiplier tube (dark rate $\simeq 30$/sec) and recorded by high-throughput photon-counting electronics operated in the delayed coincidence mode. The overall collection efficiency of the system (photoelectric counts per fluorescent photon) is estimated to be 7×10^{-8}. To acquire a transient spectrum, the Fabry-Perot is frequency stepped under control of microcomputer via a digital-to-analog converter. At each frequency step, time-resolved fluorescence signals are accumulated as the excitation cycle is repeated at a rate of 200–300 kHz with duty factor $\simeq 65\%$. Peak counting rates of approximately 100/sec were achieved at this experimental repetition rate. The computer is programmed to update a real-time display during the 1-h-long runs.

Questions of resolution are important in this experiment. The Fabry-Perot interferometer was found to have a temporal response time (e^{-1} energy decay time) of $\simeq 60$ nsec as would have been predicted on the basis of mirror reflectivity. Its spectral resolution when used to analyze scattered monochromatic laser light was 5 MHz FWHM, or roughly twice the value expected on the basis of the decay-time–determined Fourier limit.

On the other hand, the narrowest fluorescence peaks observed were 10 MHz. Much of this excess width is associated with the finite acceptance angle of the interferometer and the concomitant spread of viewing angles relative to the atomic-beam normal. (Measurements of spectrally integrated fluorescence produced by a weak laser field oriented normal to the atomic beam revealed only a 3–4-MHz Doppler width.) The product of the overall experimental spectral and temporal resolution (10 MHz and 60 nsec, respectively) is about 4 times the Fourier-transform limit. The spectral widths of all features reported below are instrumentally limited.

Figure 1(a) shows the transient spectrum of resonance fluorescence obtained for a constant-phase, square-pulse, resonant driving field of 19-MHz Rabi frequency. All Rabi frequencies quoted were measured in separate observations of optical nutation under identical experimental conditions. During the laser pulse, the spectrum consists of a symmetric, three-peaked structure similar to that observed in stead-state experiments.[13–15] Interestingly, at the end of the laser pulse, the spectrum collapses to a single component centered at the frequency of the unperturbed atomic transition.

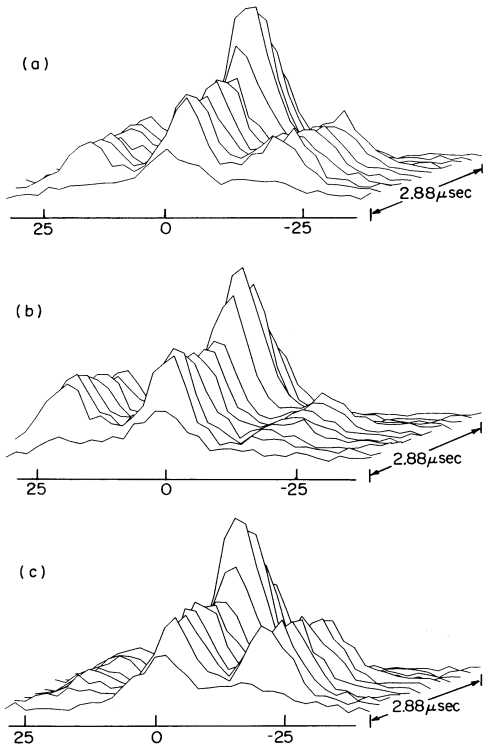

FIG. 1. Transient resonance-fluorescence spectra observed during excitation by a resonant square laser pulse of 19-MHz Rabi frequency and 1.9-μsec duration. Horizontal: $\omega_{FP}-\omega_0$, where ω_{FP} is the pass frequency of the Fabry-Perot interferometer and ω_0 is the unperturbed atomic transition frequency (all in megahertz). Vertical: Relative number of detected photons. Successive traces represent time intervals of 240 nsec. Parts (a)–(c) were obtained under identical experimental conditions except that in (a), (b), and (c), respectively, the phase of the laser driving field was constant, shifted by $+\frac{1}{2}\pi$ rad, and shifted by $-\frac{1}{2}\pi$ rad. The laser pulse begins roughly 150 nsec into the first time interval and ends during the interval in which the amplitude of the central peak increases.

Figure 1(b) illustrates the spectrum observed when a $+\frac{1}{2}\pi$-radian driving-field phase shift is applied shortly after the beginning of a resonant, square excitation pulse. As predicted,[16] the spectrum is strikingly different from its steady-state analog. It consists of a peak at the atomic transition frequency and an enhanced blue sideband. The red sideband has been almost entirely suppressed. The exact timing of the phase shift was adjusted to minimize the optical nutation observed in separate measurements of the total fluorescence. As described in Refs. 21 and 22, the effect of the phase shift is to polarize the dressed states of the atom-field system, i.e., after the phase shift only one of the levels within each dressed-state doublet is populated. As discussed elsewhere,[16] the sign of the phase shift determines whether it is the upper ($+\frac{1}{2}\pi$) or lower ($-\frac{1}{2}\pi$) component of the doublet that is left populated. As time progresses, the lower sideband begins to grow, indicating that spontaneous emission events are destroying the phase-shift–induced initial dressed-state polarization.

In Fig. 1(c), the spectrum resulting from a phase shift of $-\frac{1}{2}\pi$ is shown. In this case, one expects[16] the lower component of the dressed-state doublet to be populated. Consistent with this statement, a near perfect suppression of the blue sideband is observed early on in the spectrum.

In terms of the two-level atom vector model,[24,25] polarization of the dressed-state doublets corresponds to alignment of the Bloch vector parallel or antiparallel to the pseudofield vector.[21,22] When so aligned, the Bloch vector remains motionless and semiclassically one might expect both sidebands to disappear. The appearance of one sideband is an interesting reminder of the intrinsically quantum nature of the resonance fluorescence spectrum. In the absence of a phase shift, the Bloch vector oscillates between ground and excited atomic states indicating an amplitude modulation of the atomic dipole moment and populations. This modulation has long been used[1,5] to motivate intuitively the presence of sidebands in the Mollow spectrum.

It is interesting to note that in the case of nonresonant excitation, asymmetries in the resonance fluorescence spectrum can be induced without a phase shift. In Fig. 2, we present a transient spectrum obtained during and following a constant-phase, square, laser excitation pulse tuned 15 MHz below the atomic resonance. During the $\simeq$2-μsec laser pulse, the spectrum consists of a strong feature at the laser frequency and two sidebands. In this case, the asymmetry of the sidebands apparently results because of adiabatic following[26] effects that occur during

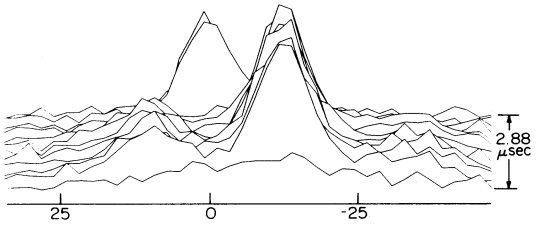

FIG. 2. Transient resonance-fluorescence spectrum observed during excitation by a detuned, square, constant-phase laser pulse of 19-MHz Rabi frequency and 1.9-μsec duration. The laser is tuned 15 MHz below resonance. Axes are as described in Fig. 1. The laser pulse begins toward the end of the first time interval.

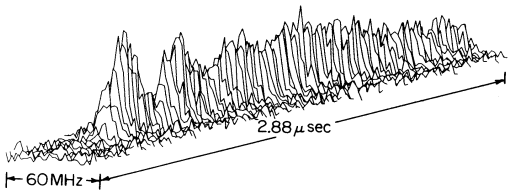

FIG. 3. Transient resonance-fluorescence spectrum observed during a relatively weak, resonant, square laser pulse of 3-MHz Rabi frequency and 1.0-μsec duration. Axes are as described in Fig. 1 except that successive traces correspond to time intervals of 20 nsec.

the nonzero laser turnon time. In the limit of a very long rise time, the Bloch vector and the pseudofield vector remain aligned, indicating complete dressed-state polarization, and one sideband should be suppressed entirely. Following the laser pulse, the atom can radiate only at its natural frequency, and the spectrum collapses to a single peak. This peak decays at a rate consistent with the decay of the upper-state population.

As mentioned above, in the absence of initial dressed-state polarization, transient resonance-fluorescence spectra are expected to oscillate in intensity as they approach their steady-state form. These oscillations are expected to occur at the effective Rabi frequency. In the spectra shown in Figs. 1(a) and 2, the predicted oscillations are too fast to be observed in our experiment because of the slow temporal response of the Fabry-Perot interferometer. In Fig. 3, we show a spectrum observed during a resonant, constant-phase, square, excitation pulse whose Rabi frequency was low enough (3 MHz) to make the temporal oscillations visible. Unfortunately, in this case the spectral resolution is insufficient to allow observation of the three spectral components expected. In order simultaneously to observe spectral intervals and temporal oscillations both determined by the Rabi frequency, it is necessary to operate the experiment in a regime such that the overall time-bandwidth product of the apparatus approaches the Fourier-transform limit. This turns out to be difficult experimentally. As mentioned above, the time-bandwidth product of the present apparatus was about 4 times larger than the transform limit.

Laser light scattered by apertures and optics in the apparatus contributes to the observed spectrum. When spectrally resolved, this scattered light artificially enhances peaks at the carrier frequency. The spectra of Fig. 1 have been generated from the raw data by subtraction of a Lorentzian-shaped background peak whose width (5 MHz FWHM) was chosen to resemble observed scattered-laser-light spectra. The magnitude of the background peak was not measured directly; therefore, its magnitude was adjusted to leave the area of the central peak in Fig. 1(a) equal to the combined area of the sidebands.[4,16-19] The same background was used in generating all parts of Fig. 1. The background deduced in this manner amounted to $\simeq$25% of the raw central-peak signal. As a check of this procedure, we note that in the reduced data of Fig. 1 the carrier feature approximately doubles in magnitude at the end of the excitation pulse as expected.

The use of extremely homogeneous excitation, optical phase and amplitude switching, a high-resolution, high-collection-power Fabry-Perot interferometer, and high-speed photon-counting electronics have made measurements of the transient spectrum of resonance fluorescence possible. Spectra from dressed-state polarized and unpolarized atoms were observed and found to be qualitatively different. Together with recent studies of atomic dynamics in the presence of a strong driving field,[22] these spectra establish a clear framework for viewing the connection between atomic dynamics and spectra.

We gratefully acknowledge financial support from the National Science Foundation under Grant No. PHY85-04260 and the U.S. Office of Naval Research under Contract No. N00014-85-K-0724, and we acknowledge stimulating interactions with N. Lu and P. R. Berman.

[1]C. Cohen-Tannoudji, in *Frontiers in Laser Spectroscopy,* Proceedings of the Les Houches Summer School, Session XXVII, edited by R. Balian, S. Haroche, and S. Liberman (North-Holland, Amsterdam, 1977).

[2]For reviews of strong-field spectra, see P. L. Knight and P. W. Milonni, Phys. Rep. **66**, 21 (1980); J. D. Cresser, Phys. Rep. **94**, 47 (1983).

[3]H. J. Kimble and L. Mandel, Phys. Rev. A **13**, 2123 (1976).

[4]B. R. Mollow, Phys. Rev. **188**, 1969 (1969).

[5]B. Renaud, R. M. Whitley, and C. R. Stroud, Jr., J. Phys. B **10** L19 (1976).

[6]S. S. Hassan and R. K. Bullough, J. Phys. B **8**, L147 (1975).

[7]M. E. Smithers and H. S. Freedhof, J. Phys. B **8**, 2911 (1975).

[8]H. J. Carmichael and D. F. Walls, J. Phys. B **8**, L77 (1975).

[9]H. J. Carmichael and D. F. Walls, J. Phys. B **9**, 1199

(1976).
[10]G. Oliver, E. Ressayre, and A. Tallet, Lett. Nuovo Cimento **2**, 777 (1971).
[11]C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977).
[12]For recent work on resonance fluorescence during pulses of well defined area, see M. Lewenstein, J. Zakrzewski, and K. Rzazewski, J. Opt. Soc. Am B **3**, 22 (1986).
[13]F. Schuda, C. R. Stroud, and M. Hercher, J. Phys. B **7**, L198 (1974).
[14]W. Hartig, W. Rasmussen, R. Schieder, and H. Walther, Z. Phys. A **278**, 205 (1976).
[15]F. Y. Wu, R. E. Grove, and S. Ezekiel, Phys. Rev. Lett. **35**, 1426 (1975); R. E. Grove, F. Y. Wu, and S. Ezekiel, Phys. Rev. A **15**, 227 (1977); M. G. Prentiss and S. Ezekiel, Phys. Rev. A **35**, 922 (1987).
[16]N. Lu, P. R. Berman, Y. S. Bai, J. E. Golub, and T. W. Mossberg, Phys. Rev. A **34**, 319 (1986).
[17]J. H. Eberly and K. Wodkiewicz, J. Opt. Soc. Am. **67**, 1252 (1977).
[18]J. H. Eberly, C. V. Kunasz, and K. Wodkiewica, J. Phys. B **13**, 217 (1980).
[19]J. H. Eberly, in *Quantum Electrodynamics and Quantum Optics,* edited by A. O. Barut (Plenum, New York, 1984).
[20]B. Renaud, R. M. Whitley, and C. R. Stroud, Jr., J. Phys. B **10**, 19 (1977).
[21]Y. S. Bia, A. G. Yodh, and T. W. Mossberg, Phys. Rev. Lett. **55**, 1277 (1985); N. Lu, P. R. Berman, A. G. Yodh, Y. S. Bai, and T. W. Mossberg, Phys. Rev. A **33**, 3956 (1986).
[22]J. E. Golub, Y. S. Bai, and T. W. Mossberg, Phys. Rev. A (to be published).
[23]M. Hercher, Appl. Opt **7**, 951 (1968).
[24]R. P. Feynmann, F. L. Vernon, Jr., and R. W. Hellwarth, J. Appl. Phys. **28**, 49 (1957).
[25]M. D. Levenson, *Introduction to Nonlinear Laser Spectroscopy* (Wiley, New York, 1984).
[26]L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).