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RAPID COMMUNICATIONS

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Phase-sensitive dynamics of bichromatically driven two-level atoms

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Observations of qualitatively different coherent transient phenomena associated with bichromatic optical excitation are reported. The effects demonstrated include the control of atomic dynamics through a variation of the initial relative phase of the driving fields and the polarization of population within atom-field dressed states. The dynamics observed are fundamentally more complex than those characteristic of monochromatically driven atoms.

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First-approximation modeling of complex systems with analogous but elementary systems is a powerful and widely employed approach to gaining insight into physical problems. For this reason, a complete and detailed understanding of the dynamics displayed by elementary systems assumes an importance far beyond that which might be accorded on the basis of the elementary system itself. In the case of light-matter interactions, it is found that a wide array of phenomenology important in science and technology can be modeled and to a large extent understood in terms of the interaction of light with simple two-level systems. In this context, it is not surprising that a tremendous body of theoretical and experimental work on driven-two-level-atom dynamics and spectra has accumulated. Much of this work has been directed toward situations involving the interaction of two-level atoms with a monochromatic driving field, and has led to the cataloging of a number of now classic atomic responses such as Rabi oscillations [1–3], adiabatic following [4], and three-peaked strong-field resonance fluorescence [5–9]. On the theoretical side, a "dressed-atom" description of the atom-field interaction has been developed [2,10] that is especially useful in quantum optical applications.

The response of the same paradigmatic two-level atom to electromagnetic fields containing more than one frequency component has received much less attention. Initial work in this area indicates that qualitatively new types of dynamical [11–17] and spectral [18–23] behavior arise. In the transient regime, for example, it has been predicted that bichromatically driven atoms exhibit dynamical behavior that is strongly dependent on the initial relative phase of the excitation-field components [11,12]. Such behavior is without analog and in fact without meaning in the case of monochromatically driven atoms and must therefore be regarded as fundamentally new phenomenology. The rich dynamics associated with bichromatically driven two-level atoms can, as in the simpler monochromatic case, be expected to provide a framework for understanding new regimes of light-matter interactions in more complex systems.

In this Rapid Communication, we report results of an experimental study of atomic dynamics in the presence of a transient bichromatic excitation field. We consider the special case wherein one bichromatic field component, the pump, is resonant with the atomic transition and the other component, the perturber, is weak. In these experiments, we demonstrate [11,12] phase-dependent dynamics and the essentially complete polarization of population within the doublets of dressed states that describe the atom coupled solely to the pump component of the bichromatic field. We refer to these eigenstates as the atom-pump dressed states. The dressed-state polarization effect may be useful in the context of manipulating dipole forces experienced by atoms [24] or in preparing atomic beams for use in injected coherence lasers [25].

Our experimental studies have focused on the transient response of two-level atoms to excitation by a step-

function bichromatic excitation field. The atoms are initially in their ground state and the initial phase difference between the two bichromatic field components is under experimental control. We assume that the two-level atom has a transition frequency ω_0, an electric-dipole matrix element d, and a wave function $\Psi = a_1|1\rangle + a_2|2\rangle$. The bichromatic field consists of a strong pump and a moderate strength perturber having frequencies ω_0 and $\omega_0 + \Delta$, respectively. We write the two-component field as

$$E(t) = \left[1/2(\varepsilon_R + \varepsilon_P e^{i(\Delta t+\phi)})e^{i\omega_0 t} + \text{c.c.}\right] S(t), \quad (1)$$

where ε_R and ε_P are real constants denoting the strengths of the two fields and $S(t) = 0$ $[S(t) = 1]$ for $t \leq 0$ $(t > 0)$.

We define the Rabi frequency of each field component as $\Omega_i = d\varepsilon_i/\hbar$, where $i = R, P$. In all of our experimental measurements, we set the perturber-field detuning Δ equal to the Rabi frequency of the resonant field Ω_R, and thereby restrict our attention to situations wherein the perturber is resonant with transitions between the atom-pump dressed states of the system. Transient atomic responses as a function of time for various values of ϕ are recorded.

A schematic of the apparatus employed to generate phase-controlled, step (10-nsec turn-on), bichromatic fields is shown in Fig. 1. A frequency-stabilized ring-dye-laser beam is passed through two acousto-optic modulators (AOM's) each driven by a bichromatic rf field. The outputs of two separate and free-running rf oscillators (one is tunable) are combined to create the bichromatic rf field. On the basis of first-order interactions, one expects two frequency-shifted optical beams to be created by the bichromatically driven AOM. In fact, higher-order interactions lead to additional nearly copropagating beams whose frequencies correspond to various combinations of the rf frequencies. The second AOM employed in our apparatus was originally intended to cancel the angular deflection of the first-order beams produced in AOM1. The presence of strong higher-order acousto-optic interactions complicated this simple picture. The deflection-cancelled output beam of AOM2 was found to contain a variety of frequency components, some of whose magnitude could be adjusted through control of the phase of the bichromatic rf field used to drive AOM2. After adjustment to eliminate unwanted spectral components, we obtained an output optical field with three components. The third spectral component was four times lower in power than the weaker of the two desired bichromatic components. Calculations indicate that the third field component has negligible impact on atomic behavior under the conditions addressed by our experiment. A pure bichromatic field is perhaps more easily obtained using monochromatically driven AOM's and optical beam combiners. In using this approach, however, preservation of phase coherence between the fields is more problematic.

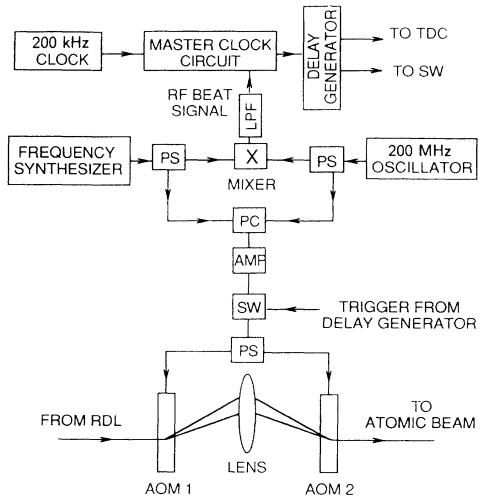

FIG. 1. Experimental-schematic. TDC: Time-to-digital converter; LPF: low pass rf filter; PS(C): power splitter (combiner); AMP: rf amplifier; SW: rf switch; RDL: ring dye laser; X: rf mixer.

Bichromatic excitation pulses were generated as follows. A 200-kHz clock enabled a master control circuit (MCC). Once enabled, the MCC was triggered by a zero crossing in the beat signal produced by mixing the two rf drive fields. Once triggered, the MCC switched on the bichromatic optical field after a programmable delay. The rf beat signal is coherent with, but phase shifted relative to, the phase difference between the optical bichromatic field components. Thus by controlling the delay between zero crossings of the rf beat signal and optical field switch on, precise shifts in the initial relative phase difference between the optical fields can be introduced. The absolute optical phase difference was not measured. Instead, it was treated as a free parameter in the theoretical modeling of the experimental results.

The experiments are performed in an atomic beam of nuclear-spin-zero ^{174}Yb atoms utilizing the $^1S_0$-$^3P_1$ transition at 556 nm. We measure the atomic fluorescence $I_f(t)$, following the onset of the step turn-on bichromatic field. $I_f(t)$ is proportional to atomic population in the excited state and hence to $[1 + R_3(t)]/2$, where $R_3(t) = (a_2 a_2^* - a_1 a_1^*)$ represents the atomic inversion. Our experimental results for various values of ϕ are presented in Figs. 2(a)–2(e). In each case, $\Omega_R/2\pi \cong 15$ MHz, $\Omega_P/2\pi \cong 1.5$ MHz, and $\Delta/2\pi \cong 15$ MHz. Fluorescence collection was controlled to ensure that contributing atoms experienced excitation-field Rabi frequencies uniform to within 2%. Atoms were in their ground state prior to excitation. In all cases [Figs. 2(a)–2(e)], $I_f(t)$ and hence R_3 display a 15-MHz oscillation. This oscillation, which represents pump-field-induced optical nutation [1] through the excited state, is damped by effects such as atomic spontaneous emission (excited-state lifetime 875 nsec) and laser field inhomogeneity. The latter effect leads to variations in Ω_R throughout the sample. The resulting distribution of nutation frequencies leads to a temporal damping of the integrated nutation signal.

For certain values of ϕ, the 15-MHz oscillation in $I_f(t)$ exhibits a deep, slow modulation [see Figs. 2(a) and 2(e)] while for other values of ϕ it does not. This modulation is not directly related to the amplitude modulation (which in our case is relatively small) intrinsically present in a bichromatic excitation field. This modulation represents a response characteristic to nonmonochromatic excitation.

To gain insight into the dynamics represented by the $I_f(t)$ curves of Figs. 2(a)–2(e), we have numerically integrated the atomic equations of motion under idealized experimental conditions (i.e., assuming zero rise-time, step, bichromatic excitation beginning at $t = 0$, spatially uniform excitation, and no atomic relaxation) for two different choices of ϕ (0 and $+\pi/2$). In both cases, $\Omega_R = \Delta$ and $\Omega_P/\Omega_R = 0.025$. We display the temporal evolution of the atom using the well-known vector model [1]. In this model, the complete atomic state is represented by a vector $\mathbf{R} = (R_1, R_2, R_3) = [a_1^* a_2 + a_2^* a_1, i(a_1^* a_2 - a_2^* a_1), a_2 a_2^* - a_1^* a_1]$. As mentioned above, R_3 represents the atomic inversion and is directly related to our experimental signal. In Fig. 3, we plot the trajectory traced by the end point of the $\mathbf{R}$ vector as it evolves under the influence of the bichromatic excitation field. When $\phi = 0$, the trajectory of $\mathbf{R}$ stays [see Fig. 3(a)] primarily in the 2-3 plane, behaving in a manner similar to that which would be expected if the perturber field were not present. The oscillation in R_3 observed in this case as $\mathbf{R}$ moves around the circular orbit at frequency Ω_R has the same properties normally ascribed to the optical nutation observed in monochromatically driven atoms. However, when $\phi = +\pi/2$, the $\mathbf{R}$ vector spirals out of the 2-3 plane [11,12], turning about a cone that slowly closes down onto the $+\hat{\mathbf{1}}$ axis [see Fig. 3(b)]. For times beyond those represented in Fig. 3(b), $\mathbf{R}$ becomes nearly aligned with the $+\hat{\mathbf{1}}$ axis, spirals back through the 2-3 plane, and then spirals down toward the $-\hat{\mathbf{1}}$ axis. This process repeats cyclically. Note that for an initial relative phase $\phi = -\pi/2$, the $\mathbf{R}$ vector will start by spiraling down toward $-\hat{\mathbf{1}}$. For $\phi = \pm\pi/2$, the spiraling process described leads to a slow modulation in the amplitude of the R_3 oscillations. It is this modulation that is observed, for example, in Fig. 2(a).

Interestingly, despite the small relative size of the perturber field it can actually dominate the evolution of the atomic state. This is not generally true. The magnitude of the perturber field's influence stems, in our experimental situation, from the fact that the perturber is resonant with a transition between the atom-pump dressed states of the system. Alternatively, in the vector model, one can view the large effect of the perturber following from a resonance between the pump Rabi frequency and the perturber detuning.

We see [Fig. 3(b)] that the perturber field can drive the atomic-state vector into alignment with either $\pm\hat{\mathbf{1}}$ axis. As has been discussed and demonstrated previously [26], this alignment corresponds to polarization of the population within the doublets of atom-pump dressed states. The cycling of the atomic-state vector between parallel and antiparallel alignment with the $\hat{\mathbf{1}}$ axis, as occurs for example in Figs. 2(a) and 3(b), is equivalent to perturber-field-induced optical nutation of population on the perturber-resonant transition between atom-pump dressed states.

Computer simulations of the expected atomic fluorescence intensity under more realistic experimental conditions are shown in Figs. 2(f)–2(j). This simulation incorporates the effects of spontaneous emission and the

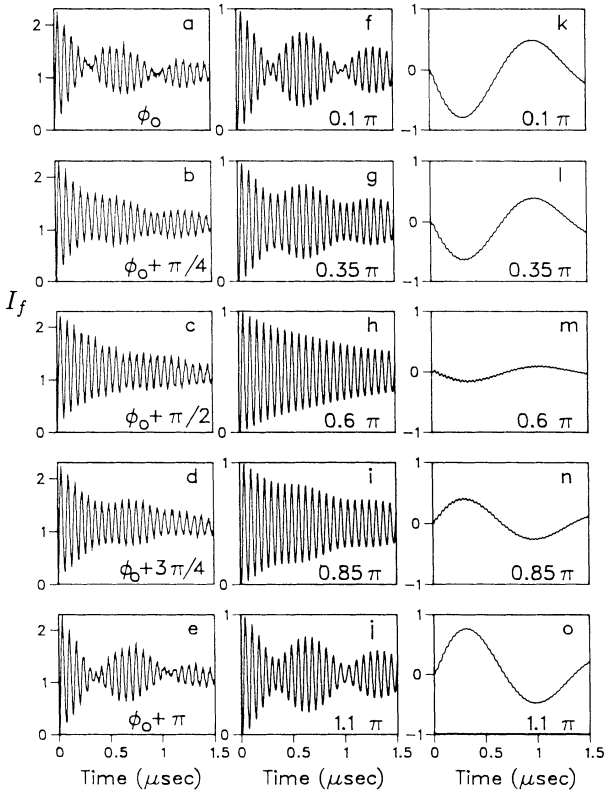

FIG. 2. (a)–(e) Experimentally measured atomic fluorescence $I_f(t)$, following onset of a step turn-on bichromatic field. Changes in the relative phase of the bichromatic field components are indicated at the lower right of each trace. (f)–(j) Calculations of $I_f(t)$ by numerical integration of the equations of motion accounting for spontaneous radiative decay and the finite rise time of the step excitation field. (k)–(o) The $\hat{\mathbf{1}}$ component of the atomic-state vector $\mathbf{R}$, derived from the same calculation as parts (f)–(j). R_1 represents the inversion of the atom-pump dressed-states.

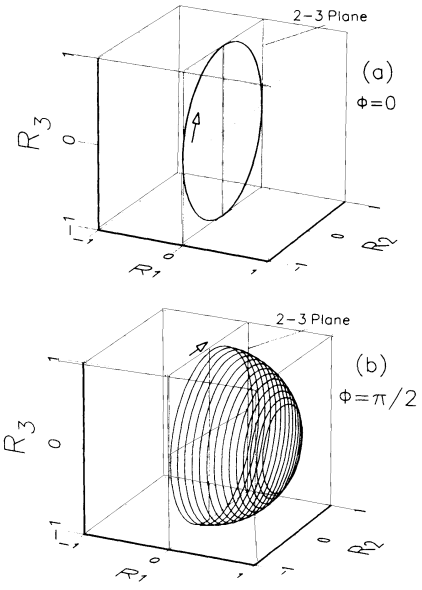

FIG. 3. Temporal evolution of atomic-state vector $\mathbf{R}$ during step bichromatic excitation commencing at t=0. Atoms are initially in their ground state, i.e., $\mathbf{R}(t) = (0, 0, -1)$. (a) $\phi = 0$; (b) $\phi = +\pi/2$.

switch-on time of the bichromatic fields, but not the residual spatial inhomogeneities of the pump and perturber fields. Interestingly, the finite switch-on time of the bichromatic field does not appear to dramatically modify the dynamical behavior associated with true step excitation as explored in Fig. 3. It introduces, rather, a shift in the values of ϕ that produce equivalent responses. The value of ϕ given in the lower right corner of Fig. 2(f) was adjusted to most accurately reproduce the data of Fig. 2(a). For pure (zero rise time) step excitation, the behavior of Fig. 2(f) corresponds to $\phi \approx 0.5\pi$. The ϕ values used in Figs. 2(g)–2(j) were shifted from the Fig. 2(f) value in accordance with experimentally introduced phase shifts.

In Figs. 2(k)-2(o), we plot the projection of the atomic-state vector $\mathbf{R}$ onto the $\hat{\mathbf{1}}$ axis versus time. Parameters utilized correspond to those of Figs. 2(f)–2(j). This projection represents the atom-pump dressed-state inversion. It exhibits a modulation whose amplitude is ϕ dependent and whose frequency is $\Omega_P/2$. In Figs. 2(k) and 2(o), corresponding to the excitation conditions of Figs. 2(a) and 2(e), respectively, the dressed-state inversion approaches unit magnitude at the times that the high-frequency modulation of $I_f(t)$ vanishes. The disappearance of optical nutation when the dressed states are polarized has been demonstrated previously. [26]

One of the more interesting aspects of the transients demonstrated here is their dependence on the initial phase difference between the two bichromatic field components. In the case of monochromatic excitation, atomic responses are also phase dependent *if the interaction begins with the atom in a superposition state.* In the present experiments, the atoms begin in their ground state. Importantly, however, the atomic ground state corresponds to a superposition of atom-pump dressed states. The phase dependence observed in the present instance arises because the perturber field resonantly drives an atom-pump dressed-state transition and atomic-ground-state preparation corresponds to preparation of the atom-pump states in a superpostion state.

In conclusion, we have demonstrated that atomic dynamics in the presence of transient bichromatic excitation are qualitatively different from those seen in the case of monochromatic excitation. We find that such differences can persist even when one of the bichromatic field components is arbitrarily weak. We have pointed out the perturber's impact can be understood in our case because of its resonance with transitions between atom-pump dressed states. We have shown that the initial relative phase of the bichromatic field components plays a vital role in determining the atomic response, and that certain initial relative phases lead to complete atom–pump-field dressed-state polarization. Knowledge of these different classes of behavior may contribute to the understanding of dynamical behavior in a wide range of circumstances and provide new tools for the control of atom-field behavior.

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