Selective Excitation of Dressed Atomic States by Use of Phase-Controlled Optical Fields

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We have studied the response of atoms in a dilute collimated atomic beam to transient phasecontrolled resonant excitation fields. We demonstrate that the use of such excitation makes it possible to place the atoms into stationary states of the coupled atom-field system. We show that these states, which are easily visualized in the two-level-atom vector model, are equivalent to the dressed states of resonance fluorescence theory. Studies of atoms in pure dressed states should allow interesting tests of basic atom-field interactions.

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The interaction of simple two-level quantum systems (atoms) with nearly resonant electromagnetic excitation fields has been a subject of great interest over the years. Perhaps the most elegant description of the dynamics involved is afforded by the two-level-atom vector model.^{1,2} In the vector model, the atom and driving field are represented by vectors in a threedimensional space, and the atomic response to the driving field is simply visualized as a precession of the atomic-state vector about the instantaneous drivingfield vector. In the case of a constant driving field, two orientations of the atomic-state vector become very special: the atomic-state vector becomes motionless when it points parallel or antiparallel to the driving-field vector. This implies that there is no energy exchange between the excitation field and the atom even if the atom and the field are exactly resonant. The special atom-field states can thus be described as stationary states, and as discussed below, they correspond precisely to the "dressed" states familiar from the theory of resonance fluorescence.³⁻⁵ As such they should have interesting spectral properties. In this Letter, we describe the first experimental observation of stationary atom-field states involving resonant optical excitation fields. These special states were generated through the use of amplitude- and phase-controlled excitation of a collimated atomic beam. We note that stationary spin-field states were studied in nuclear magnetic resonance work, where they were referred to as "spin-locked" states.¹

Consider the response of a two-level atom [see Fig. 1(a)] to an oscillating electric field of the form

$$E(t) = \mathscr{E}_0 \cos(\omega t + \varphi), \tag{1}$$

where \mathscr{C}_0 and φ are constants. With use of the rotating-wave and electric-dipole approximations, and expressed in a frame rotating at the laser frequency ω , the atom-field interaction Hamiltonian is given by

$$\hbar \begin{pmatrix} 0 & -\chi^* \\ -\chi & \Delta \end{pmatrix}, \tag{2}$$

where $\chi = p \mathscr{C}_0 \exp(-i\varphi)/2\hbar$, and *p*, assumed real, is

the appropriate electric dipole moment. The eigenvalues and eigenstates associated with this Hamiltonian are

$$\lambda_{+} = \frac{1}{2}\hbar \left[\Delta \pm (\Delta^{2} + p^{2} \mathscr{C}_{0}^{2}/\hbar^{2})^{1/2} \right], \tag{3a}$$

and

$$W_{\pm}(\varphi) = \alpha_{\pm} \left(\frac{-p\mathscr{C}_0}{2\lambda_{\pm}} \exp(i\varphi) \psi_a + \psi_b \right), \qquad (3b)$$

respectively, where $\alpha_{\pm} = [(p^2 \mathscr{C}_0^2 / 4\lambda_{\pm}^2) + 1]^{-1/2}$, and ψ_a and ψ_b are eigenstates of the isolated atom. Note that the eigenvectors but not the eigenvalues depend on the instantaneous phase of the excitation field. The eigenvectors W_{\pm} are the semiclassical dressed atomic states discussed elsewhere.^{6,7} When field quantization is taken into account,³⁻⁵ the dressed states become linear combinations of atomic and photon states. In terms of energy, the fully quantized dressed states form an extended series of doublets.³⁻⁵ When one semiclassical dressed state is populated, one state within *each* doublet of fully quantized dressed states is populated.

The atomic-state vector, ρ , used in the vector



FIG. 1. (a) Two-level atom and relevant frequencies. (b) Electric field amplitude and phase vs time. The phase switch at time t_2 was not always employed.



FIG. 2. (a) Evolution of ρ during the interval $t_1 > t > t_0$, with $\theta_{10} = \pi/2$ and $\varphi_0 = 0$. (b) Stationary atom-field state for $t > t_1$, with $\varphi_1 = \pi/2$. (c) Same as (b) but with $\varphi_1 = -\pi/2$.

 $model^2$ is given by

$$\boldsymbol{\rho} = \{(ab^* + ba^*), i(ba^* - ab^*), (bb^* - aa^*)\}, \quad (4)$$

where a and b are the expansion coefficients of the instantaneous atomic state in terms of ψ_a and ψ_b , respectively. The atomic-state vector obeys the wellknown equation

$$d\rho/dt = \omega(\varphi) \times \rho, \tag{5}$$

where $\boldsymbol{\omega}$, the driving-field vector, is given by

$$\boldsymbol{\omega}(\varphi) = \left[\frac{-p\mathscr{E}_0}{\hbar}\cos\varphi, \frac{-p\mathscr{E}_0}{\hbar}\sin\varphi, \Delta\right]. \tag{6}$$

The physical significance of the dressed states is appreciated by calculation of their corresponding atomic-state vectors. It is found that $W_{\pm}(\varphi)$ corresponds to the atomic-state vector

$$\boldsymbol{\eta}_{\pm}(\varphi) = \boldsymbol{\beta}_{\pm}\boldsymbol{\omega}(\varphi), \tag{7}$$

where

$$\beta_{\pm} = \hbar \{ \lambda_{\pm} [(p^2 \mathscr{C}_0^2 / 4 \lambda_{\pm}^2) + 1] \}^{-1}.$$
(8)



FIG. 3. Schematic of experiment. RDL, ring dye laser; BD, beam dump; AOM, acousto-optic modulator; EOM, electro-optic modulator; A1, rectangular aperture; P, polarizer; $\lambda/4$, quarter-wave plate; L, lens; and OB, optical baffle.

The dressed states, W_{\pm} , are seen to correspond quite generally to atomic-state vectors, η_{\pm} , which lie parallel (+) and antiparallel (-) to the instantaneous driving field. By simply designing an excitation sequence which aligns ρ and ω , one can selectively populate a single dressed state. This applies to both resonant and nonresonant excitation. This intuitively appealing result is implicit in earlier work,⁶ but its general significance does not appear to have been appreciated.

In our experiment, we expose two-level atoms in a collimated atomic beam to a phase-controlled resonant excitation field like that shown in Fig. 1(b).⁸ We monitor the state of the atoms by observing their time-dependent resonance fluorescence. Since our atomic beam is dilute, and we observe at right angles to the laser propagation direction, the fluorescence intensity, $I_F(t_m)$, at any arbitrary measurement time, t_m , should be directly proportional to the excited-state population. Straightforward calculations reveal that

$$I_F(t_m) = I_0 \sin^2(\theta_{m0}/2) \quad (t_1 > t_m > t_0), \tag{9a}$$

and

$$I_F(t_m) = I_0 \{ \cos^2(\theta_{10}/2) \sin^2(\theta_{m1}/2) + \cos^2(\theta_{m1}/2) \sin^2(\theta_{10}/2) + [\sin(\theta_{10}) \sin(\theta_{m1}) \cos(\varphi_1 - \varphi_0)]/2 \}$$
(9b)
(t_2 > t_m > t_1),

where I_0 is a constant, $\theta_{ij} = p \mathscr{C}_0 t_{ij}/\hbar$, $t_{ij} = t_i - t_j$, and φ_i (i = 0, 1) represents the phase of the excitation field as shown in Fig. 1(b). These expressions depend on time implicitly through the angles θ_{m0} and θ_{m1} . In general, $I_F(t_m)$ is oscillatory; however, if $\theta_{10} = (r + \frac{1}{2})\pi$ and $|\varphi_1 - \varphi_0| = (s + \frac{1}{2})\pi$, where $r, s = 0, 1, 2, \ldots$, the time dependence of I_F vanishes. A moment's reflection reveals that these values of θ_{10} and $|\varphi_1 - \varphi_0|$ are precisely those that lead to alignment of ρ and ω for $t > t_1$ (see, for example, Fig. 2). We use the absence of oscillations in the fluorescence intensity to indicate that we have successfully aligned ρ and ω . In the interval before the phase shift is applied, we simply see

Rabi oscillations similar to those seen in optical nutation experiments.^{9,10} Of course, in the present case, we observe fluorescence rather than coherent forward scattering.

Our experiment (see Fig. 3) was performed on the 555.6-nm $6s^{21}S_0$ - $6s6p^3P_1$ transition of atomic ¹⁷⁴Yb. Having zero nuclear spin, ¹⁷⁴Yb constitutes a nearly ideal two-level system when excited by circularly polarized light. The lifetime of the excited state is a convenient 875 nsec. A commercial ring dye laser actively locked to its standard reference cavity provided the excitation field which was amplitude gated with acousto-



FIG. 4. Effect of a $\pi/2$ phase shift applied at successively later times on the fluorescence intensity vs time (see text).

optic modulators and phase controlled with an electrooptic modulator. High-voltage power metal-oxidesemiconductor field-effect transistors were employed to introduce variable-amplitude phase shifts with switching times of ≈ 8 nsec. To ensure a very uniform laser intensity throughout the laser-Yb interaction region, a 1-mm-square aperture (A1) was positioned to pick out the uniform-intensity central region of the laser beam, and two well-corrected doublet lenses imaged (at unity magnification) the aperture into the atomic beam. Laser power in the interaction region was about 10 mW. The beam contained all the natural isotopes, but they could be easily separated spectrally. Scans of the ¹⁷⁴Yb absorption profile revealed a 5-MHz residual beam Doppler width. The effects of stray magnetic fields in the laser-Yb interaction region were minimized by applying a \sim 6-G magnetic field, coaxial with the circularly polarized laser field. Short-focal-length Fresnel lenses provided a



FIG. 6. Effect of phase shifts of various magnitudes on the fluorescence intensity vs time (see text).

fluorescence collection efficiency of about 10%. The excitation sequence was repeated at approximately 10 kHz, and fluorescence intensity was recorded with a gated boxcar integrator. A complete scan of the fluorescence intensity versus time for a particular excitation sequence required about 100 sec. Longer scan times were precluded by laser frequency drift. The atomic beam was run at rather low density; only about 1000 atoms were presented in the interaction region during a particular laser pulse.

In Figs. 4–6, we show recordings of the fluorescence intensity, I_F , versus time during various excitation sequences of the form shown in Fig. 1(b). In Fig. 4, the effect of a fixed $\pi/2$ phase shift $(\varphi_1 - \varphi_0)$ applied at successively larger values of t_{10} (and hence θ_{10}) is examined. In trace (i), there is no phase shift (equivalent to $t_{10}=0$), and I_F should ideally display the simple oscillatory behavior characteristic of Eq. (9a). The observed damping arises primarily from residual laser-intensity inhomogeneity in the interaction region, but other effects, such as transit time, natural decay, and residual Doppler broadening, also contribute. In the remaining traces, (ii) through (x), the phase shift occurs farther and farther into the pulse. Arrows below several traces indicate the approximate time at which the phase shift was applied. Note the



FIG. 5. Reduction of dephasing in stationary atom-field states (see text).

flatness of the curve of I_F versus time after the phase shifts in traces (iv) and (viii). In these traces, $\theta_{10} = \pi/2$ and $3\pi/2$, respectively, and as expected, stationary atom-field states have been produced. Traces (vi) and (x) are also interesting because the I_F curve is essentially unaffected by the phase shift. These traces correspond, respectively, to $\theta_{10} = \pi$ and 2π , which implies that the phase shift is applied when the atoms are purely in one atomic state.

In Fig. 5, we show that dephasing can be inhibited in the special aligned states. Trace (i) shows dephasing with no phase shift. In trace (ii), a $\varphi_1 - \varphi_0 = \pi/2$ phase shift is applied with $\theta_{10} = \pi/2$ and is left on for the 500-nsec interval indicated. When the field's phase is returned to its initial value, at time t_2 [see Fig. 1(b)], I_F still displays a relatively large oscillation. Note that this oscillation is much larger than that shown at earlier times in trace (i). Since the stationary atom-field states lie in the 1-2 plane of the Bloch sphere, conventional wisdom asserts that they should decay with the transverse dephasing time T_2 . In our system, natural decay alone makes $T_2 \cong 1750$ nsec. Preliminary measurements show the delayed signal to decay at a rate roughly consistent with the effects of natural decay and beam transit time; however, more detailed studies of this problem are planned.

Finally, in Fig. 6, we set $\theta_{10} = \pi/2$ and vary $|\varphi_1 - \varphi_0|$. The traces, from top to bottom, represent phase shifts of 0, $\pi/2$, and π radians, respectively. The dashed vertical line represents the time at which the phase shift was applied. Note that after a phase shift of π , the fluorescence intensity decreases, i.e., the atoms are deexcited by the field.

In summary, we have demonstrated the generation of stationary atom-field states, and identified these states with the dressed states of resonance fluorescence theory. More generally, we have demonstrated the capability of studying samples in which all constituent atoms have been prepared in essentially identical and arbitrary initial states. It will thus be possible to test current understanding of effects such as resonance fluorescence under quite general conditions.

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