Observation of Dressed-Atom Effects in Three-Level Free-Induction Decay

H. W. H. Lee[a] and J. E. Wessel

Chemistry and Physics Laboratory, The Aerospace Corporation, El Segundo, California 90254

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New coherent transient optical effects are reported for resonantly excited three-level cesium atoms. Complex modulation of free-induction decay was observed and is attributed to dressed-atom effects. Contrary to prior expectations, coherent transients monitored for upper and lower transitions were remarkably dissimilar and provide clear evidence for coherence transfer between the upper and lower optical transitions.

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Free-induction decay (FID) is a fundamental optical process which was extensively investigated and applied in earlier work on laser interactions involving two-level systems. In the FID process, the off-diagonal polarization of an atomic ensemble generates a coherent electromagnetic wave. Typically, it is detected by heterodyne means through interference between the emitted wave and the forward-propagating exciting wave. This can strongly modify the temporal characteristics of a transmitted laser beam. The resultant signal contains a rich variety of information about the radiative interaction, including atomic population and coherence-dephasing processes.

Whereas coherent radiative interactions of two-level systems are well understood, currently there is considerable interest in excitation processes of three-level systems. Lu *et al.*[1] recently elucidated the important dynamical consequences of dressed-atom state preparation on three-level transient spectra. Experimental results for ytterbium in an atomic beam[2] were in complete agreement with a fully developed theoretical treatment.[1] Their work extended prior energy-domain studies of Autler-Townes interactions in three-level systems into the time domain.

In this Letter, new results are presented from experimental and theoretical investigation of three-level FID in resonantly excited three-level cesium atoms using the dc-Stark-shifting technique.[3] We observed dramatic amplitude modulation of FID signals, and established that this arises from dressed-atom effects. Furthermore, in contrast to earlier studies by Loy[4] and Liao, Bjorkholm, and Gordon,[5] we observed the FID of both the lower and the upper transitions and found appreciable differences between the two. These observations provide the first clear evidence for simultaneous FID and nutation in the lower transition, induced by a Stark shift of the upper energy level of the upper transition.

Several years ago studies were initiated on FID in two-photon absorption in three-level systems. The nonresonant two-photon coherent processes were found to be well described by a simple two-photon Bloch-equation treatment.[3,4] In contrast, preliminary studies of resonant two-photon FID in sodium, performed by Liao, Bjorkholm, and Gordon[5] using the Stark switching technique originally introduced by Brewer and Shoemaker[3] for two-level systems, revealed interesting FID signals with phases dependent both on detuning of the first beam (ω_1) and on intensity of the second beam (ω_2). This effect was not understood, prompting Marshman *et al.*[6] to study the problem theoretically. On the basis of results obtained from a simplified density-matrix model, they postulated that the π phase change in the signal arises from a population inversion. It is interesting that Marshman *et al.* modeled the upper transition, whereas Liao, Bjorkholm, and Gordon studied the lower transition. At that time this was probably not considered significant, because prior work on Raman beats by Brewer and Hahn and others,[7] and by Shoemaker and Brewer,[8] suggested that the resonant two-photon excitation of a three-level atom is essentially identical to the three-level Raman-beat experiment, for which the ω_1 and ω_2 beams are modulated similarly. We demonstrate below that though this may hold for a three-level system in an inverted "V" configuration (where the final level is lower in energy than the intermediate level, giving rise to a Raman-like process), this is not the case for a three-level system in a ladder configuration (where each successive level is higher in energy).

Our experiments were performed with a heated (145 °C) cesium cell equipped with two 10-cm-long parallel electrodes spaced by 0.4 cm. Excitation was provided by two counterpropagating single-longitudinal-mode ring-dye-laser beams (with optical frequencies ω_1 and ω_2) tuned to excite the $6\,^2S_{1/2}$-$6\,^2P_{3/2}$ transition (852 nm) and the $6\,^2P_{3/2}$-$7\,^2D_{5/2}$ transition (698 nm) (with Rabi frequencies χ_1 and χ_2). The relevant energy levels and excitation scheme are shown in Fig. 1(b). The cesium density was such that both the ω_1 and ω_2 beams penetrated the cell without strong attenuation. A dc voltage of about 5 kV/cm was applied to the electrodes in order to isolate spectrally the $7\,^2D_{5/2}(m = \pm\frac{1}{2})$ component. A square pulse, of at most 300-V amplitude and 100-ns duration, propagated down the traveling-wave electrode structure, shifting the more polarizable

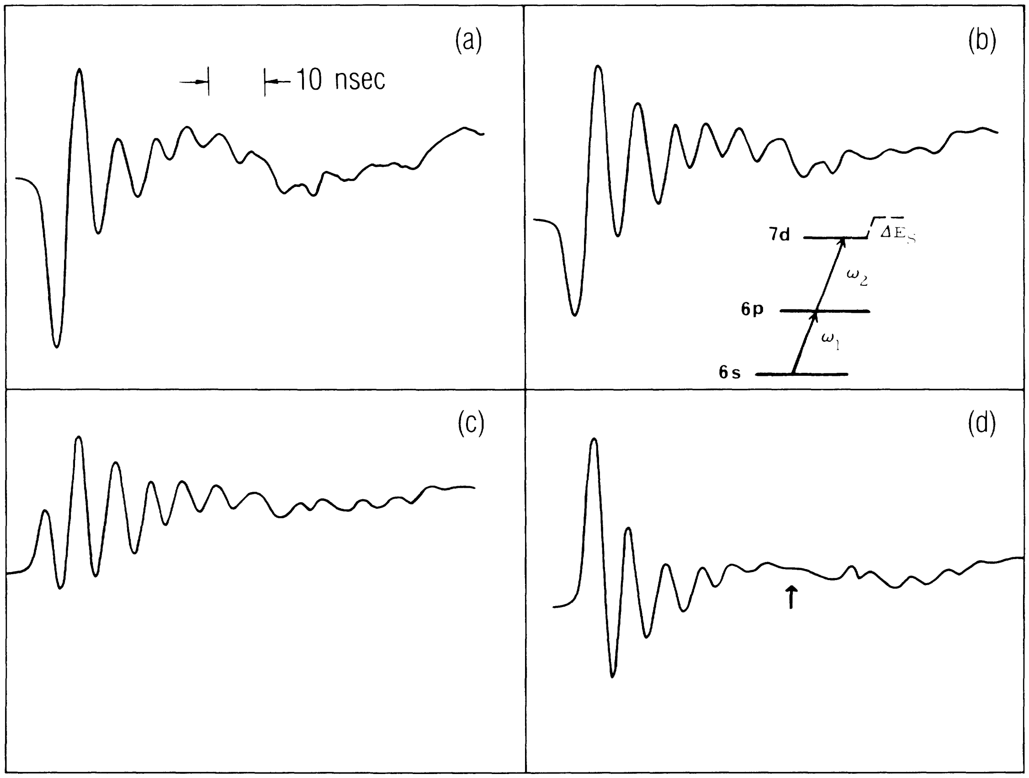

FIG. 1. FID signal observed by the monitoring of the transmitted ω_2 beam as a function of ω_2 detuning. The Stark shift is 140 MHz. Recorded (a) at +40-MHz detuning, (b) on resonance, (c) at −40-MHz detuning, and (d) at −80 MHz detuning.

$7\,^2D_{5/2}(m=\pm\frac{1}{2})$ component up to 160 MHz (ΔE_S) and leaving the $6\,^2P_{3/2}$ component essentially unshifted.[9] The P-polarized beams were of 0.3-cm diameter and the power levels were maintained at about 50 mW in each beam. Beams emerging from the cell were focused onto individual photodiodes, which had response times of an order of magnitude less than the fastest recorded features of the signals. The data presented here were recorded with a fast−rise-time amplifier together with a boxcar averager.

Figure 1 shows the FID signals observed in the upper transition (ω_2) as a function of detuning of the ω_2 probe beam. The waveforms are far more complex than the simple damped sinusoidal waveforms which are typical for two-level systems. The amplitudes of successive oscillation peaks vary erratically and the period is not constant, particularly for resonant excitation conditions. Structure in the complex modulation patterns is strongly dependent on the ω_1 intensity. This previously unobserved behavior is interpreted below as resulting from dressed-atom effects.

Signals imposed on the ω_1 beam were also monitored, as shown in Fig. 2(b). Contrary to initial expectations,[7] they are remarkably different from the ω_2 signals [Fig. 2(a)]. The rapidly oscillating FID has a significant slow nutation component. This nutation is dependent on χ_2 and is highly dispersive in ω_2, unlike an ordinary nutation. Surprisingly, the ω_1 signals are modulated at the frequency of the Stark shift of the upper state ($7d$), even though the upper state is not directly involved in the ω_1 transition. The extent of modulation and the phase are dependent on ω_2 resonance conditions. These results differ dramatically from prior observations of FID and nutation.

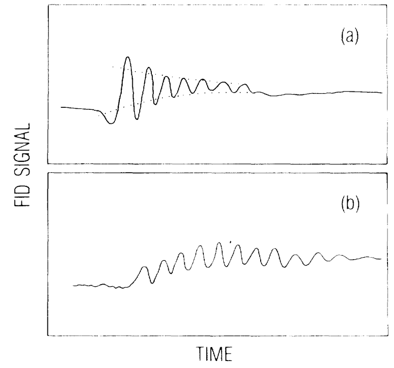

FIG. 2. Comparison between the FID observed in (a) the upper (ω_2) transition and (b) the lower (ω_1) transition. The Stark shifts are (a) 160 MHz and (b) 140 MHz; full scale is 90 ns in both cases.

In many other respects the three-level FID signals have a close relationship to behavior previously observed for two-level systems. The heterodyne beat frequency observed in the ω_1 and ω_2 transitions at low χ_2 is close to that expected on the basis of the 140-MHz Stark shift of the final level. In Fig. 1 it is clear that the ω_2 FID envelopes are highly nonexponential, with fast and slow decay contributions. This is particularly obvious in the resonant case shown in Fig. 2(a), where the Stark shift was increased to 160 MHz. The fast and slow decays correspond, respectively, to the linear and nonlinear FID described by Brewer and co-workers[10] for a two-level inhomogeneously broadened system. The linear FID is dispersive in nature and accounts for the π phase shift observed Fig. 1. This represents the first observation of linear FID in a three-level system. In contrast, the nonlinear FID is absorptive and decays with a slower exponential envelope [described by the dotted curves in Fig. 2(a)], with a rate determined by the power-broadened width.

As noted in Ref. 10, the linear FID for a two-level atom escaped notice for some time as a result of its rapid decay (typically subnanosecond). However, the analogous linear FID for a three-level atom, shown in Figs. 1 and 2(a), is readily observable, decaying in about 10 ns. The order-of-magnitude difference is a result of the reduced effective inhomogeneous width in resonance with ω_1 and the additional Doppler reduction due to the counterpropagating configuration. This is completely consistent with the observed decay rate of the linear FID.

The above data were modeled by rigorous density-matrix calculations. Two approaches were used. One was performed analytically with use of the bare-atom representation, and the other approach was based on the dressed-atom representation.[1] The analytical results were Doppler averaged numerically and the dressed-atom calculations were performed entirely numerically. Results were similar for the two approaches, within the range of validity for the numerical results. The two-photon coherence contribution to $\mathrm{Im}\rho_{23}$ was immediately evident in the analytical expressions. Equally apparent was the biexponential decay of $\mathrm{Im}\rho_{23}$, in contrast to the single exponential of two-level systems. In addition, the solution shows the apparent dephasing rate to be different from what would be expected from a simple application of two-level concepts to a three-level system. The additional contributions to the dephasing rate are dependent on the laser detunings, the Stark shift, and the Rabi frequency χ_1. There are also additional contributions to the Stark-shift–induced beating frequency in the FID. The detailed mathematical description will be presented in a future publication.

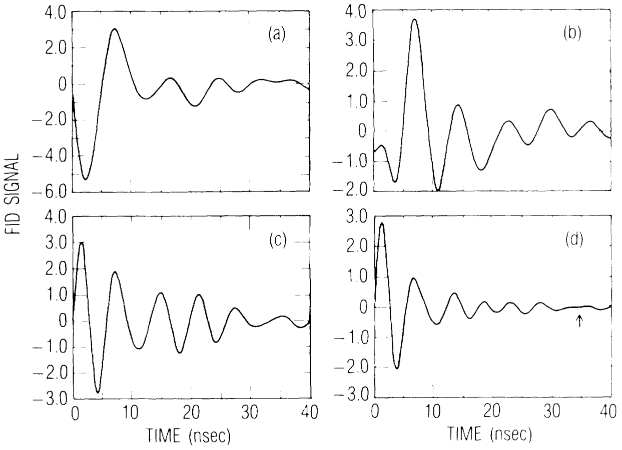

FIG. 3. Calculated FID of the upper transition (ω_2) as a function of ω_2 detuning. The Stark shift is 140 MHz. (a) +40-MHz detuning, (b) on resonance, (c) −40-MHz detuning, and (d) −80-MHz detuning. The arrow in (d) represents cancellation between FID contributions from the two lower-transition dressed-atom states.

Figure 3 presents results calculated by numerical Doppler averaging of the analytically integrated bare-atom density-matrix equations. The FID signal for the ω_2 beam is presented for various ω_2 detunings, at a fixed ω_1. The Stark shift of the final level was 140 MHz and the Rabi frequencies for the lower (χ_1) and upper (χ_2) transitions were 121 and 40.7 MHz, respectively. As expected from simple extrapolation from two-level results, the FID oscillations dampen rapidly with increased ω_2 detuning. More importantly, the calculations reproduce the observed phase shift as the line center is crossed. This arises from dispersion, not from population inversion, as previously proposed.[6] The rigorous calculation also duplicates the observed difference in oscillation frequency between the linear and nonlinear FID components, and correctly predicts the survival of only the linear FID with large detunings. There is excellent agreement between the calculated FID amplitude envelope and the observed erratic FID oscillation amplitudes.

Calculations were carried out for larger Stark-shift frequencies. The greater temporal resolution of the FID envelope aided in our elucidating the complex amplitude modulation of the FID envelope. An example is shown in Fig. 4(a), where the Stark shift is 2 GHz, χ_1 is 85.9 MHz, and χ_2 is 40.7 MHz. The unusual FID envelope is dramatically evident for large Stark shifts. Zero positions of the modulation envelope are strongly dependent on χ_1.

The origin of the FID amplitude modulation is readily apparent from the dressed-atom description. We performed a second set of calculations based on the dressed-atom treatment of Ref. 1, using a numerical integration. This calculation confirmed that the modulation envelope arises from interference between two separate FID terms, originating from the two dressed-atom states prepared by the ω_1 interaction. Analytical

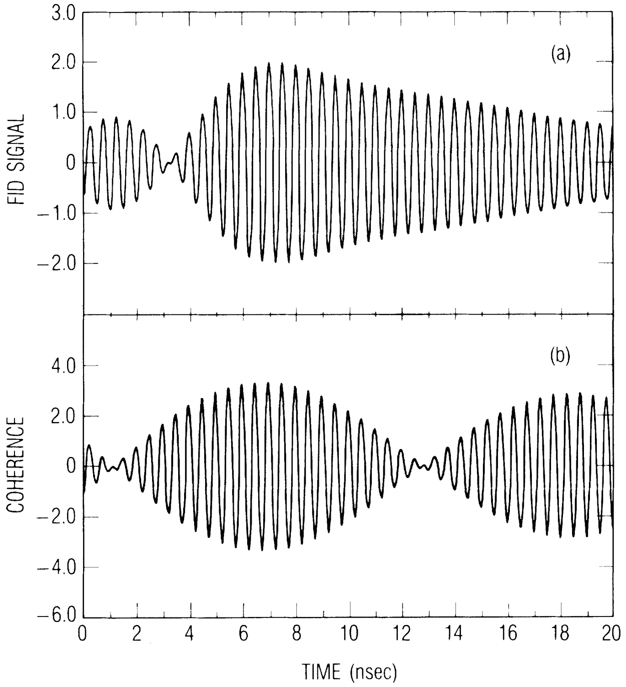

FIG. 4. (a) Calculated FID of the upper (ω_2) transition with a 2-GHz Stark shift. The Doppler width is suppressed in (b) in order to reveal the FID modulation introduced by interference between FID contributions from the two lower-transition dressed-atom states split by a χ_1 of 85.9 MHz.

results are shown in Fig. 4(b) where Imρ_{23} is displayed for a Rabi frequency for the lower transition, χ_1, of 85.9 MHz. The coherence Imρ_{23} is clearly amplitude modulated at χ_1, the dressed-atom state splitting. Additional calculations confirm that the amplitude modulation of Imρ_{23} varies with χ_1. This effect is dramatically manifested in the experimentally observed FID in Fig. 1(d), where the arrow indicates a point of complete cancellation in the interference of the FID from the two dressed-atom states. The behavior is reproduced by the calculation shown in Fig. 3(d).

Our dressed-atom calculation also predicted signals resembling those observed for the ω_1 beam. Calculations and the observed ω_2 dependence imply that the slowly varying component of the signal is associated with nutation, indicating that population transfer is an important component of the three-level FID process. The ω_1 signal is also weakly modulated at the Stark-shift frequency of the upper state, and the overall wave form varies, depending on experimental conditions, from nearly pure FID resembling the ω_2 signal to a dispersive nutation-type signal, with weak modulation at the Stark frequency.

In conclusion, the new results show how dressed-atom effects are strongly manifested in the FID of three-level systems. The major differences between FID observed in the upper and the lower transitions demonstrate that coherence processes of three-level systems are more subtle than previously recognized. Coherence transfer between these two transitions is readily observable and can be distinguished from stepwise incoherent population transfer. On the basis of these results, we believe that FID will provide a sensitive method to monitor the effects of atomic collision processes on coherent interactions involving dressed states.

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[a] Present address: Lawrence Livermore National Laboratory, Livermore, CA 94550.

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