RESONANCE FLUORESCENCE DURING PHASE-CONTROLLED TRANSIENT EXCITATION

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The response of a two-level atom to a resonant driving field can be dramatically altered by a sudden shift in the driving field's phase. We discuss the implications of this fact with respect to the atom's transient fluorescence spectrum and relaxation properties.

Extensive work [‡1] [1–5] over the last ten to twenty years has led to a fairly detailed understanding of the triple-peaked spectrum of steady-state, strong-field resonance fluorescence. Some work [6,7] has also been directed toward elucidating the properties of transient resonance fluorescence, but to date, only a very limited range of transient situations has been considered. In the present report, we discuss the effect of novel initial conditions on the transient fluorescence problem. We point out that a particular phase-controlled excitation scheme leads to a *transient* fluorescence spectrum in which one of the Rabi sidebands normally observed in steady-state situations is initially suppressed. This excitation scheme has been discussed in the context of nuclear magnetic resonance (NMR), and is associated with the process known as "spin locking" [8]. Aside from its effect on transient fluorescence spectra, the optical analog of spin locking should be interesting because of its ability to suppress relaxation processes [8].

We focus our attention on experiments which might be performed with the apparatus shown in fig. 1. We assume that the beam of two-level atoms is perfectly collimated so that the effects of inhomogeneous broadening can be neglected. We also assume that the laser excites the atoms exactly at their single-photon transition frequency ω_0, and that the atomic beam is sufficiently dilute that all collective effects can be neglected. Fluorescent light is observed in a direction orthogonal to that of both the atomic and laser beams.

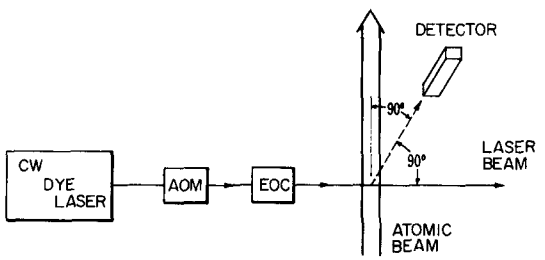

Fig. 1. Hypothetical apparatus for the study of transient resonance fluorescence. The dye laser is assumed frequency stabilized, continuous wave, and single mode. The AOM (acousto-optic modulator) turns the laser field in the laser–atomic-beam interaction region on and off. The EOC (electro-optic crystal) is used to vary the phase of the laser field. The detector is mounted normal to both the atomic and laser beams.

First, consider the implementation of one of the transient resonance fluorescence experiments discussed by Eberly et al. [6]. In this experiment, the acousto-optic modulator (AOM) is suddenly switched on, and fluorescence from the initially unexcited atoms is observed. A vector model [‡2] description of the atomic behavior in this situation is shown in fig. 2. Viewed from a frame co-rotating with the driving field $\boldsymbol{\Omega}$, the atomic pseudo-spin vector, $\boldsymbol{\rho}$, precesses in the 1–3 plane. This situation is actually quite similar to

[‡1] For a relatively recent review see ref. [4].

[‡2] We use the notation of ref. [9].

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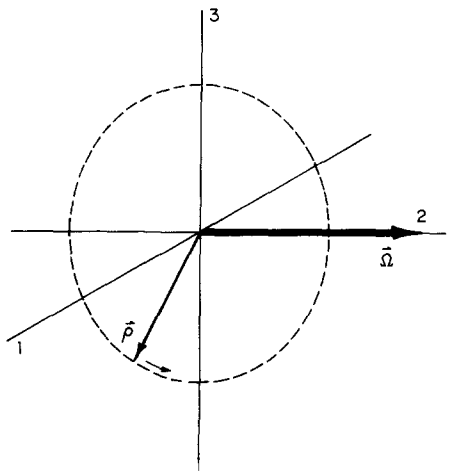

Fig. 2. Evolution of the pseudo-spin vector $\boldsymbol{\rho}$ (in a frame rotating at the laser frequency) after a steady laser field is switched on. The atom is assumed to have been initially in its ground state. As $\boldsymbol{\rho}$ precesses in the 1–3 plane, its transverse component (i.e. the component in the 1–2 plane) is amplitude modulated. The atom's electric dipole moment is proportional to the transverse component of $\boldsymbol{\rho}$.

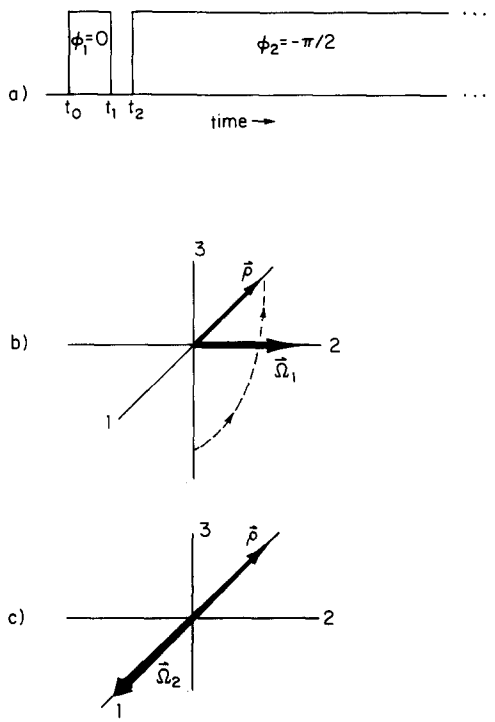

Fig. 3. (a) Laser field amplitude as a function of time. ϕ_i gives the relative phase of the laser field during the ith pulse. (b) Evolution of the rotating-frame, pseudo-spin vector, $\boldsymbol{\rho}$, during the interval $t_0 < t < t_1$ of part (a). $\boldsymbol{\rho}$ is shown in its final position at $t = t_1$. For $t < t_0$, the atom is assumed to be in its ground state. (c) Spin-locked situation found for $t > t_2$. The pseudo-vector, $\boldsymbol{\rho}$, remains motionless regardless of the magnitude of $\boldsymbol{\Omega}_2$.

that found in the steady-state case, except of course for the fact that here all the atoms in the sample precess in unison. In view of this similarity, it is not particularly surprising to note that the calculated transient spectrum [6] displays a triple-peaked structure closely resembling that calculated for the steady-state case [1–4].

The pseudo-spin behaves quite differently in a spin-locking experiment. In this case (see fig. 3a), the atoms are first prepared by a laser pulse having a duration $t_{10} = t_1 - t_0$ and an effective driving field $\boldsymbol{\Omega}_1$. The pulse parameters are controlled so that $|\boldsymbol{\Omega}_1| t_{10} = \pi/2$. The laser field places the initially unexcited atoms into an equal admixture superposition of their ground and excited states (see fig. 3b). When $t = t_1$, the laser field is turned off and a voltage is applied to the electro-optic crystal (EOC) so that its optical path length changes by one quarter of a wavelength. The interval $t_{21} = t_2 - t_1$ is assumed very short compared to all relaxation processes. When $t = t_2$, the laser field is again switched on. Because of the electro-optically induced optical path change, the atom–field relative phase is shifted by $-\pi/2$ radians (the minus sign is chosen for convenience) and the driving field, $\boldsymbol{\Omega}_2$, and the pseudo-spin vector, $\boldsymbol{\rho}$, are antiparallel (see fig. 3c). As a result, the pseudo-spin vector remains fixed in position regardless of the strength of the applied optical field. Using the terminology of NMR [8,10], the pseudo-spin vector in fig. 3c is said to be "spin-locked" by the driving field $\boldsymbol{\Omega}_2$.

The two spin-locked states ($\boldsymbol{\rho} \parallel \pm \boldsymbol{\Omega}_2$) are special. They are the dressed states often employed in the description of resonance fluorescence [3,4]. By controlling the sign of ϕ_2 (see fig. 3), one can selectively populate either of the two dressed states. The dressed-state population imbalance created through spin-locking is thermalized in the course of spontaneous radiative decay.

It is interesting to consider the time-dependent spectrum [6,7] of the resonance fluorescence emitted by a sample of spin-locked atoms. Immediately after the phase shift one dressed state is unpopulated. A

standard dressed-state analysis [3,4] predicts that the resulting fluorescence spectrum will then contain only two out of the three spectral peaks characteristic of steady-state resonance fluorescence, i.e. one of the sidebands usually observed will be absent. By changing the sign of ϕ_2, one can control which sideband is suppressed. As the atoms fluoresce, the dressed states become equally populated and the initially suppressed sideband should reappear. Measurements of the time-dependent fluorescence spectrum [6,7] made within the first radiative lifetime after spin-locking has occurred should be able to resolve this behavior. We note that other means of selectively exciting particular dressed states have been discussed [4,11], and concepts related to time-dependent spectra in three-level systems analyzed [12].

It is also interesting to consider the interaction of a spin-locked atom with a stochastic noise field $\boldsymbol{\Omega}_n(t)$. Both the amplitude and phase of $\boldsymbol{\Omega}_n(t)$ may vary randomly in time. We restrict our attention to situations in which $|\boldsymbol{\Omega}_n|\tau_c \ll 1$, where τ_c is the correlation time associated with the noise field. In this case, the noise field causes the atom to undergo a diffusive type relaxation. As discussed in works on NMR [8,10], when $|\boldsymbol{\Omega}_2|\tau_c \gg 1$, the pseudo-spin vector adiabatically follows the total driving field $\boldsymbol{\Omega}_T = \boldsymbol{\Omega}_2 + \boldsymbol{\Omega}_n$ as it jitters around the direction of $\boldsymbol{\Omega}_2$. As a result, the applied field is highly effective in suppressing noise-field-induced diffusion of the pseudo-spin vector away from its initial locked position. If the condition $|\boldsymbol{\Omega}_2|\tau_c \gg 1$ is not satisfied, the pseudo-spin diffuses away from its locked position at essentially the same rate as would be observed with no locking field.

One might wonder whether a locking field of moderate intensity could possibly suppress the rate of spontaneous radiative decay. Since spontaneous radiative decay is often viewed as resulting from a noise field associated with vacuum fluctuations and/or radiation reaction [13], this is a reasonable question to ask. This noise field is normally assumed to have a very short ($\approx 1/\omega_0$) correlation time [3], thus conventional wisdom asserts that *extremely* large optical fields should be necessary to perturb spontaneous fluorescence rates. The spontaneous emission noise field cannot, however, be completely analogous to a classical noise field, since the latter leads to spontaneous absorption as well as emission. One is led to speculate that the downward directed diffusion characteristic of spontaneous radiative decay might be affected by relatively weak locking fields. This speculation can be tested in optical spin-locking experiments.

We have recently demonstrated [14] optical spin locking in a system similar to that shown in fig. 1, and we are currently studying the various properties discussed here.

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