

**Suppression and coherent control of free-induction-decay emission in multilevel systems**

Luís E. E. de Araujo\*

*Instituto de Física “Gleb Wataghin,” Universidade Estadual de Campinas, Campinas, São Paulo 13083-970, Brazil*

(Received 6 July 2006; published 19 October 2006)

In this paper, I study the coherent control and suppression of the free-induction-decay emission associated with the decay from an excited multilevel atom to its ground state. It is shown here that a strong and ultrashort coupling pulse, resonant with the excited states and a lower state other than the ground state, can induce destructive quantum interferences in the decay process. The suppression is temporary and the free-induction-decay signal reappears one quantum-beat period (of the excited states) later. A sequence of equally spaced, ultrashort coupling pulses can control the moment the free-induction-decay emission occurs.

DOI: [10.1103/PhysRevA.74.043415](https://doi.org/10.1103/PhysRevA.74.043415)

PACS number(s): 42.50.Md, 32.80.Qk, 42.50.Gy

**I. INTRODUCTION**

Quantum interference can significantly alter an atom's response to an optical field. It can suppress the absorption of a resonant laser beam, as in electromagnetically induced transparency [1], as well as enhance this absorption, as in electromagnetically induced absorption [2]. But quantum interference can also affect an atom's spontaneous decay. External fields can be used to control and suppress the atom's spontaneous emission by creating superpositions of excited states that are stable against this kind of decay [3–10].

Control of spontaneous decay has attracted much attention due to its potential application in lasing without inversion, quantum information and computing, and precision spectroscopy, for example. Zhu and Scully [3] studied the cancellation of spontaneous emission due to the interference between the decay processes of two nondegenerate upper levels to a common lower level in an open V atom excited by a single laser. Paspalakis and Knight [4] showed, in a similar kind of system, the coherent control of spontaneous emission by adjusting the relative phase of the two lasers used for the excitation. Selective and total cancellation of fluorescence, as well as extreme line narrowing, were predicted. Later, Paspalakis and co-workers [5] demonstrated an alternative scheme in which interfering paths for spontaneous decay are induced by a microwave field applied between the excited states. Control of the fluorescence spectrum in a three-level  $\Lambda$  system by driving the lower doublet with a coherent microwave field has also been proposed [6]. Evers and Keitel [7] investigated the slowdown of spontaneous emission in a two-level atom by a possibly intense field. By applying a series of ultrashort  $2\pi$  pulses to an atom, Agarwal and co-workers [9] demonstrated the inhibition of spontaneous decay from a single excited state to a continuum. In a different context, the control of spontaneous emission has also been demonstrated by placing the atom near a photonic band edge [11] and in a microcavity [12]. Control of the collective spontaneous emission of a pair of atoms has also been proposed [13].

In a multilevel system with an initial atomic polarization (induced, for example, by an ultrashort optical pulse whose bandwidth is large enough to overlap several excited states of the system), a free-induction-decay (FID) signal can be observed. The FID signal consists of rapid field oscillations that are a result of interference among the various coherent polarizations in the medium. If a large number of states are excited by the ultrashort pulse, these rapid oscillations are well localized in time, forming a sequence of spontaneously emitted coherent impulses separated in time by the quantum-beat period of the excited states [14–17]. These impulses are associated with the motion of the nonstationary superposition of the excited states, or wave packet. An impulse is generated whenever there is a wave packet recurrence; that is, the wave packet completes a revolution in the excited potential. If the excited-state spacing is much larger than the polarization decay rate (such as is the case for the Rydberg levels of atoms and the vibrational states of molecules), then many (hundreds of thousands for the mentioned systems) FID impulses will be emitted before the atomic polarization starts decaying.

In recent work [18], I have suggested that quantum interferences to a common ground state can momentarily suppress the FID emission associated with wave packet recurrences in a multilevel atom. By applying a strong and ultrashort coupling field to the atom at the moment a recurrence should occur, the atom can be momentarily prevented from emitting. The emission is delayed by one beat period of the excited states. In Ref. [18], suppression of FID emission was analyzed by numerically integrating the Maxwell-Bloch propagation equation for one particular test pulse. In this paper, I analyze that phenomenon in more detail, demonstrating its coherent nature. The Maxwell-Bloch equations are solved analytically for arbitrary pulse shapes, and I show that the control pulse induces a destructive quantum interference in the atom's free-induction-decay process. Coherent control of the emission process is demonstrated by a sequence of ultrashort coupling pulses, which controls the moment the free-induction decay occurs.

The control of emission from a superposition of excited states was also studied by Frishman and Shapiro [10]. They showed that the spontaneous decay from a manifold of excited states could be inhibited by the application of short

\*Electronic address: [araujo@ifi.unicamp.br](mailto:araujo@ifi.unicamp.br)

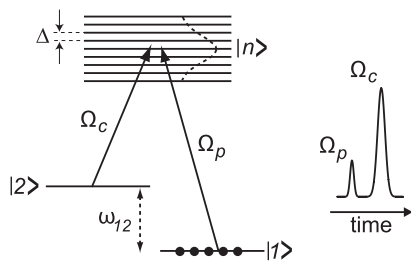


FIG. 1. A “multi- $\Lambda$ ” system excited by a pair of coupling and probe pulses delayed in time by an integer multiple of the quantum-beat period  $T=2\pi/\Delta$ . Both pulses are resonant with the same excited state and have Rabi frequencies  $\Omega_c$  and  $\Omega_p$ . The pulses have spectra (dashed line) large enough to overlap several states in the excited manifold. At the same time, their spectra are much smaller than the energy difference  $\omega_{12}$  of the lower levels. Energy separations and pulse amplitudes are not drawn to scale; solid circles represent the initial population distribution; and  $\Delta$  is the level spacing.

microwave  $\pi$  pulses. The microwave pulses, operating directly in the manifold and coupling the excited states, suppressed the decay by inducing an interference within the manifold itself, instead of between transitions to a common ground state. In their multilevel atomic system, the excited-state spacing was only five times the decay rate of the excited states. This spacing allowed them to observe “photon bursts” on the time scale of the spontaneous decay. And only an infrequent application of the microwave pulses was necessary to suppress the decay.

As I will show, for the atomic system described in this paper, the orders-of-magnitude difference between the excited state’s spacing and their decay rate prevents a similar approach (infrequent application of control pulses) for suppressing FID emission. The control pulses have to be applied every beat period of the excited states for suppression, otherwise the FID emission returns.

Figure 1 shows the atomic system considered here. It consists of two lower states and a manifold of equally spaced excited states. The equal spacing between the excited states is assumed for simplicity: in this limit, the Maxwell-Bloch propagation equations can be solved analytically. In this “multi- $\Lambda$ ” medium, a weak probe pulse connects state  $|1\rangle$  to the excited manifold of states  $|n\rangle$ , while a stronger coupling pulse connects state  $|2\rangle$  to the same excited states. The pulses are delayed in time with respect to each other, and the coupling pulse enters the medium after the probe pulse. The durations of the input pulses to be considered here are in the femto to picosecond scale, and therefore many orders of magnitude shorter than any relaxation time of the medium. Their bandwidths are large enough to overlap several of the excited states, but at the same time, they are much smaller than the energy splitting  $\omega_{12}$  of the lower levels. Applying an ultrashort probe pulse to this system creates a wave packet in the excited manifold. This wave packet oscillates in the excited potential with a characteristic time  $T=2\pi/\Delta$ —where  $\Delta$

is the level spacing—corresponding to the quantum-beat period between the excited states. The wave packet motion manifests itself as a sequence of emitted secondary impulses (the FID signal) separated in time by  $T$ , as shown in Fig. 2(a).

Many real atomic systems have part of their structure of the type shown in Fig. 1. For example, the Cs atom: starting from the  $7s$  electronic state (excited from the ground state with dye lasers), an ultrashort pulse with a central wavelength of 780 nm can excite a wave packet centered at the  $n=100p$  Rydberg state. A coupling ultrashort pulse with a central wavelength of  $1.02\ \mu\text{m}$  would then connect the  $8s$  state to the upper manifold of states, which is approximately harmonic. Although both pulses have large bandwidths, the  $7s$  and  $8s$  states are spaced far enough apart that neither pulse can alone connect both energy levels. The quantum-beat period for this system is  $T\approx 160$  ps, while the lifetime of the excited Rydberg states is on the order of microseconds. It does not matter if states  $|1\rangle$  and  $|2\rangle$  are “ground” (metastable) states or not. Everything (excitation and FID emission) happens in such a short time scale that decay from these states is not relevant; state  $|2\rangle$  may be an excited state. Then, instead of the multi- $\Lambda$  system of Fig. 1, one could equally explore a cascade configuration of electronic states of a molecule, such as the  $X\ ^1\Sigma_g^+ \rightarrow A\ ^1\Sigma_u^+ \rightarrow 2\ ^1\Pi_g$  ladder in the potassium dimer, for example. States  $|1\rangle$  and  $|2\rangle$  are then vibrational states in the ground  $X\ ^1\Sigma_g^+$  and excited  $2\ ^1\Pi_g$  electronic states, respectively; the manifold of excited states are the vibrational states of the intermediate  $A\ ^1\Sigma_u^+$  state of the cascade system. The intermediate  $A$  state has a vibrational period of  $T\approx 470$  fs, and transitions between this state and the other two electronic states can be excited by ultrashort pulses centered at 840 nm [19].

## II. THE MAXWELL-BLOCH PROPAGATION EQUATIONS

Propagation of the input probe and coupling pulses in an extended collection of the atoms of Fig. 1, along with propagation of the FID signal, is modeled by solving the Maxwell-Bloch equations. In the rotating wave approximation, the temporal evolution of the probability amplitudes  $a$ ,  $c$ , and  $b_n$  of the bare  $|1\rangle$ ,  $|2\rangle$ , and  $|n\rangle$  states, respectively, is found by solving Schrödinger’s equation [18]:

$$\dot{a}(z,t) = 0.5i\Omega_p f^*(z,t) \sum_n b_n(z,t) e^{-i\delta_n t}, \quad (1a)$$

$$\dot{c}(z,t) = 0.5i\Omega_c g^*(z,t) \sum_n b_n(z,t) e^{-i\Delta_n t}, \quad (1b)$$

$$\dot{b}_n(z,t) = 0.5i[\Omega_p f(z,t)a(z,t)e^{i\delta_n t} + \Omega_c g(z,t)c(z,t)e^{i\Delta_n t}]. \quad (1c)$$

The summations are carried over the number of levels in the excited manifold of states. The atom is initially in ground state  $|1\rangle$ , and the probe pulse is weak enough that the ground-state probability amplitude remains nearly unperturbed throughout the medium:  $a(z,t) \approx 1$ . Assuming uniform electric-dipole moments for transitions between the lower and the excited states accessed by the pulses' spectra, then  $\Omega_p$  and  $\Omega_c$  are the real Rabi frequencies associated with the probe and coupling fields, respectively, and  $\Omega_c \gg \Omega_p$ .  $f(z,t)$  and  $g(z,t)$  are the dimensionless complex amplitudes of the probe and coupling fields, respectively; and they are slowly varying (envelope as well as phase). In the problem to be addressed here,  $f(0,t)$  describes the input driving pulse, while  $f(z,t)$  represents the total probe field—driving pulse and FID impulses—in the medium. The input probe pulse  $f(0,t)$  occurs during the time interval  $0 \leq t < T$ , while  $g(0,t)$  is delayed with respect to  $f(0,t)$  by an integer multiple of the beat period  $T$ . Both  $f(z,t)$  and  $g(z,t)$  are normalized to unity at the entrance to the medium.

The detuning between the probe's carrier frequency and level  $n$  is  $\delta_n = \omega_n - \omega_p$ , with  $\omega_n$  being the eigenfrequency of level  $n$ . And  $\Delta_n = \omega_n - \omega_c$  is the detuning between the coupling field's carrier frequency and level  $n$ . For equally spaced levels,  $\delta_n = \bar{\delta} + \Delta(n - \bar{n})$ , where  $\bar{\delta} = \omega_{\bar{n}} - \omega_p$  is the probe's detuning from level  $\bar{n}$ , the average quantum number of the states excited by the probe pulse. Similarly,  $\Delta_n = \bar{\Delta} + \Delta(n - \bar{n})$ , with  $\bar{\Delta} = \omega_{\bar{n}} - \omega_c$ .

Equations (1) are to be solved along with the reduced propagation equation for the probe field:

$$\frac{\partial}{\partial z} [\Omega_p f(z,t)] = 2i\mu \sum_n P_n(z,t), \quad (2)$$

where  $P_n(z,t) = [b_n(z,t) \exp(-i\delta_n t)] a^*(z,t)$  is the atomic polarization induced in the medium by the probe field between the ground and the excited state  $|n\rangle$ ;  $\mu = \omega_L N d^2 / \epsilon_0 \hbar c$  is the coupling constant between the field and the polarization;  $N$  is the number density; and  $\epsilon_0$  is the permittivity of the multilevel medium for nonresonant transitions. In Eqs. (1) and (2),  $t$  is the local time.

Since state  $|2\rangle$  starts out unpopulated, and it essentially remains this way at all times for a weak excitation of the excited manifold, the polarization between state  $|2\rangle$  and the excited states is negligible. The coupling pulse propagates unchanged in the medium:  $g(z,t) \approx g(0,t)$ .

### III. SUPPRESSION OF THE FID SIGNAL

The FID signal is directly related to the induced polarization between the ground and excited states. From Eq. (2), canceling the atomic polarization suppresses FID emission. The goal of this section is to show that  $P(z,t) \equiv \sum_n P_n(z,t) = 0$  in the presence of a strong coupling pulse.

In the weak excitation regime, integrating Eq. (1c) gives

$$b_n(z,t) \approx 0.5i \int_{-\infty}^t ds [\Omega_p f(z,s) e^{i\delta_n s} + \Omega_c g(0,s) c(z,s) e^{i\Delta_n s}]. \quad (3)$$

Assuming a large number of states in the excited manifold (although only some of them are populated by the probe pulse), the Poisson sum formula [20] can be used to show that

$$\sum_n e^{-i\Delta(n-\bar{n})(t-s)} \approx T \sum_{m=-\infty}^{\infty} \delta(t-s-mT). \quad (4)$$

The contribution of levels with large detunings is cut off by the rapidly oscillating exponentials, and the summation limits in Eq. (4) could be extended to  $\pm\infty$  [17].

If the coupling pulse is delayed by one period  $T$  from the input probe pulse, it will coincide with the moment the first FID impulse would be emitted by the atom. Then, for times  $T \leq t < 2T$ , combining Eqs. (3) and (4), Eq. (1b) becomes

$$\begin{aligned} \dot{c}(z,t) = & -\frac{1}{4} \Omega_c T g^*(z,t) \left( \Omega_p e^{-i\bar{\Delta}t} e^{i\bar{\delta}(t-T)} f(z,t-T) \right. \\ & \left. + \frac{1}{2} \Omega_c g(0,t) c(z,t) \right). \end{aligned} \quad (5)$$

The above equation can be solved exactly for  $c(z,t)$ , but a much simpler approximate solution can be found by assuming  $\dot{c}(z,t) \approx 0$ . This should be a very good approximation in the weak excitation regime when the probability amplitude of state  $|2\rangle$  should not vary significantly. From Eq. (5)

$$c(z,t) = - (2\Omega_p / \Omega_c) \frac{f(z,t-T)}{g(0,t)} e^{-i\theta} e^{-i\vartheta t}, \quad (6)$$

where  $\theta \equiv 2\pi\bar{\delta}/\Delta$  and  $\vartheta \equiv \bar{\Delta} - \bar{\delta}$ . In the above equation,  $f(z,t-T)$  represents the driving probe pulse evaluated in the  $T \leq t < 2T$  interval. If the coupling and probe pulses have the same temporal shape, then after the coupling pulse turns off, a small population will be left in the lower state  $|2\rangle$ :  $|c(z,t)|^2 \approx 4(\Omega_p / \Omega_c)^2$ . This is the situation considered in Ref. [18]. However, Eq. (6) implies that if the coupling pulse is longer than the probe, the ratio  $f(z,t-T)/g(0,t) \rightarrow 0$  at the end of the coupling pulse, and no population will be left in state  $|2\rangle$ . At any other time, the  $(2\Omega_p / \Omega_c)$  factor guarantees that only a very small probability amplitude will be temporarily transferred to the  $|2\rangle$  state, satisfying the assumption that  $c(z,t)$  does not vary appreciably. Equation (6) is valid as long as the coupling pulse is shorter than one period  $T$ .

Substituting Eqs. (3) and (4) into  $P(z,t) \approx \sum_n b_n(z,t) e^{-i\delta_n t}$ , it is straightforward to show that

$$P(z,t) = \frac{i}{2} T \left( \Omega_p f(z,t-T) e^{-i\theta} + \frac{1}{2} \Omega_c g(0,t) c(z,t) e^{i\vartheta t} \right) \quad (7)$$

for times  $T \leq t < 2T$ . And from Eq. (6)

$$P(z,t) = 0. \quad (8)$$

Thus, a strong coupling pulse applied at the time of the first wave-packet recurrence cancels the total polarization between the ground and excited states, suppressing the FID emission associated with that recurrence. The coupling pulse modifies the phase of the excited states' probability amplitudes such that quantum interference between these states and the ground state is of a destructive nature. This result was obtained for a coupling pulse delayed by one period  $T$  from the driving pulse, but it can be easily generalized to longer delays that are integer multiples of  $T$ . This analysis can also be extended to a sequence of coupling pulses, equally separated in time by one period  $T$ , showing that the moment of FID emission can be controlled.

#### IV. PROPAGATION OF THE FID SIGNAL

When one of the FID impulses is suppressed by the coupling pulse, the propagation dynamics of the following impulses is modified. In Ref. [18], Eqs. (1) and (2) were solved numerically for a coupling pulse matched in shape to the input probe pulse, but delayed by one period  $T$  from it. The numerical solution showed that the FID signal was strongly suppressed in the  $T \leq t < 2T$  interval, and the first observed FID impulse occurred during  $2T \leq t < 3T$ . This temporally shifted impulse had a propagation dynamics that resembled that of the first FID impulse in the no-coupling-pulse case. Here, to determine how propagation is affected, Eqs. (3) and (4) are substituted into the reduced wave equation for the probe field [Eq. (2)], which becomes

$$\begin{aligned} \frac{\partial}{\partial z} f(z, t) = & -\frac{1}{2} \alpha [f(z, t) + (\Omega_c / \Omega_p) g(0, t) c(z, t) e^{i\vartheta t}] \\ & - \alpha \sum_{m=1}^{\infty} [f(z, t - mT) e^{-im\theta} \\ & + (\Omega_c / \Omega_p) g(0, t - mT) c(z, t - mT) e^{-im\phi} e^{i\vartheta t}], \end{aligned} \quad (9)$$

where  $\alpha = \omega_p \mathcal{N} d^2 T / \epsilon_0 \hbar c$  is the propagation constant and  $\phi \equiv 2\pi \bar{\Delta} / \Delta$ .

In the absence of a coupling pulse ( $\Omega_c = 0$ ), Eq. (9) can be solved analytically for any of the secondary FID impulses [17]. The input probe pulse decays exponentially with propagation distance:

$$f(z, t) = f(0, t) e^{-\alpha z / 2}, \quad (10)$$

while the first impulse in the FID signal is given by:

$$f(z, t) = -(\alpha z) e^{-\alpha z / 2} e^{-i\theta} f(0, t - T); \quad (11)$$

$f(0, t - T)$  is the input probe pulse shifted to the  $T \leq t < 2T$  time interval. The first FID impulse has the same temporal profile as the input probe pulse, but a different  $z$  dependence. Tuning of the probe's center frequency sets the relative phase between the first impulse and the input pulse: if the input pulse is tuned to a resonance ( $\theta = 0$ ), the FID impulse will be  $\pi$  rad out of phase with the input probe; if the input is tuned in between two resonances ( $\theta = \pi$ ), the FID impulse will be in phase with the input. Reference [17] lists analytical solutions for some of the following FID impulses.

A coupling pulse applied in the  $T \leq t < 2T$  interval, will not change the propagation of the input probe pulse. Solving Eq. (9) for  $\Omega_c \neq 0$  in the  $0 \leq t < T$  interval yields Eq. (10) again. Having solved for the probability amplitude  $c(z, t)$  for  $T \leq t < 2T$  in the last section, finding a analytic solution for  $f(z, t)$  in the  $2T \leq t < 3T$  time interval is straightforward. Since in this interval  $g(0, t) = g(0, t - 2T) = 0$  (the coupling pulse exists only for  $T \leq t < 2T$ ), and from Eq. (8),  $f(z, t - T) \approx 0$  [here,  $f(z, t - T)$  is not the driving probe pulse that appears in Eq. (6), but the suppressed impulse of the previous time interval], then integrating Eq. (9) gives

$$f(z, t) = (\alpha z) e^{-\alpha z / 2} e^{-2i\theta} f(0, t - 2T). \quad (12)$$

This FID impulse has the same  $z$  dependence as the first FID impulse observed in the  $\Omega_c = 0$  case [Eq. (11)]. Temporally, it is identical (in envelope and phase) to the input probe pulse. However, the relative phase between the FID impulse and the input pulse is not the same as that observed in Eq. (11): the FID impulse will be in phase with the input for both on-resonance and between-resonance tuning of the input probe pulse. Tuning of the coupling pulse has no effect on the FID impulse.

Analytical solutions for the FID impulses that follow can be obtained in a similar manner. It is found that when the probe pulse is on resonance with an excited transition, the FID impulses are  $\pi$  rad out of phase with the corresponding FID impulses of the no-coupling-pulse case. And when the probe pulse is tuned between two resonances, the FID impulses are in phase with those impulses observed with no coupling pulse present.

#### V. NUMERICAL RESULTS

The results of the previous sections were checked by numerically solving Eqs. (1) and (2) with the following test input-probe pulse:

$$f(0, t) = \exp[-(4 \ln 2)(\tau - 0.5T)^2 / a^2 + i\varphi(\tau - 0.5T)], \quad (13)$$

where  $a = 0.15T$  is the pulse's full width at half maximum. The probe's Rabi frequency was set so that its area was  $\Omega_p \int_{-\infty}^{\infty} |f(0, \tau)| d\tau = 0.1\pi$ . With such a pulse width and Rabi frequency, a numerical solution of the complete Maxwell-Bloch equations (1) and (2) shows that the spectrum of the probe pulse overlaps about 13 states in the excited manifold, out of a total of 29 excited states.

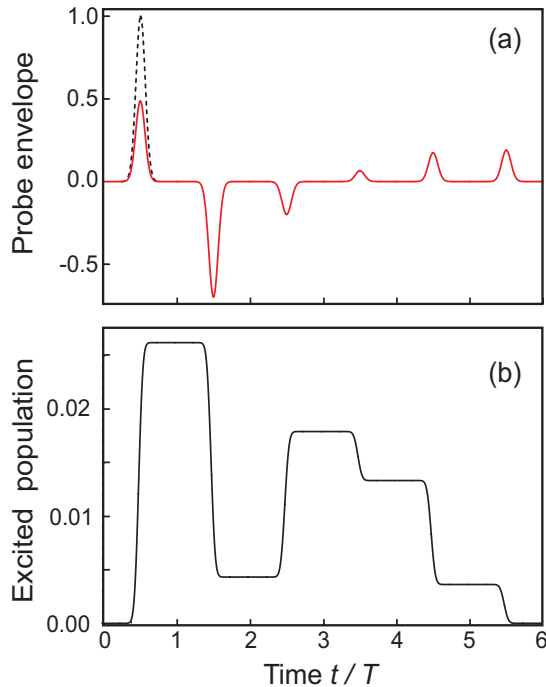


FIG. 2. (Color online) (a) Envelope  $f(z,t)$  of the probe pulse at  $\alpha z=0$  (dashed line), given by Eq. (13), and after propagating a distance of  $\alpha z=1.5$  (solid red line). Here,  $\varphi(0)=0$ . As described in the text, the input Gaussian pulse is followed by a sequence of coherent FID impulses spontaneously emitted by the atoms. (b) The total population in the excited manifold of states at  $\alpha z=1.5$ .

Figure 2(a) shows the total probe field (solid line), at a distance  $\alpha z=1.5$  inside the atomic medium, as a function of time, when the coupling pulse has not been applied. The input probe pulse (dashed line) occurs in the  $0 \leq t < T$  interval, and was assumed to be transform limited. This far into the medium, its peak amplitude has decreased to approximately 50% of its value at  $\alpha z=0$  due to absorption. The input pulse is followed by a series of FID impulses. It was set on resonance with an excited state ( $\theta=0$ ), so the first FID impulse, occurring at  $t=1.5T$ , is  $\pi$  rad out of phase with it. Figure 2(b) shows the total population in the excited manifold of states. The probe pulse transfers approximately 2.7% of the ground-state population to the excited states. (At the input,  $\alpha z=0$ , the probe pulse excites about 10% of the ground-state population.) This population then cycles between the ground and the excited states as the FID impulses are emitted.

The field inside the medium, in the presence of a Gaussian coupling pulse with twice the width of the input probe pulse and centered at  $t=1.5T$ , is shown in Fig. 3(a). Here,  $\Omega_c=50\Omega_p$ . FID emission is suppressed in the  $T \leq t < 2T$  interval, when the coupling pulse is present. The first FID impulse to occur is delayed to  $2T \leq t < 3T$ , and it is inverted with respect to that of Fig. 3(a), as predicted by Eq. (12). Figure 3(b) shows the total population in the excited state (solid line), along with the population in state  $|2\rangle$  (dashed blue line). As assumed in the previous sections, no significant amount of population is removed from the excited states to the lower state  $|2\rangle$  by the coupling pulse. A negligible

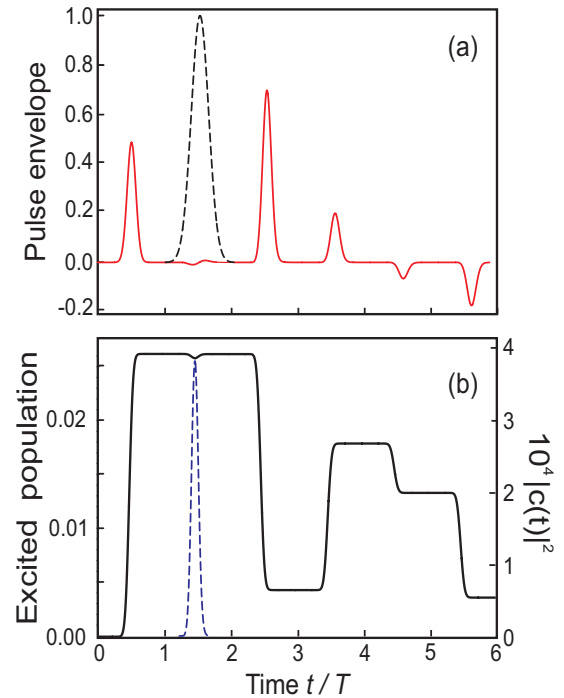


FIG. 3. (Color online) (a) Envelope  $g(0,t)$  of the coupling pulse (dashed line) and that of the probe pulse  $f(z,t)$  (solid red line). FID emission at  $t=1.5T$  is suppressed by the coupling pulse. The following FID impulses are inverted with respect to those of Fig. 2(a). (b) The total population in the excited states (solid line) and that in the lower state  $|2\rangle$  (dashed blue line). Here,  $\Omega_c=50\Omega_p$  and  $\alpha z=1.5$ .

population is placed in state  $|2\rangle$ , but by the end of the coupling pulse, it is removed back to the excited states. A peak population of less than 0.04% is observed in Fig. 3(b), in agreement with Eq. (6) when one considers that, at  $\alpha z=1.5$ , the peak amplitude of the pulse at  $t=0.5T$  has decayed to half its input value.

To check that the temporal phase of the FID impulses is preserved upon emission suppression, I considered an input test pulse identical to that of Eq. (13), but with an arbitrary

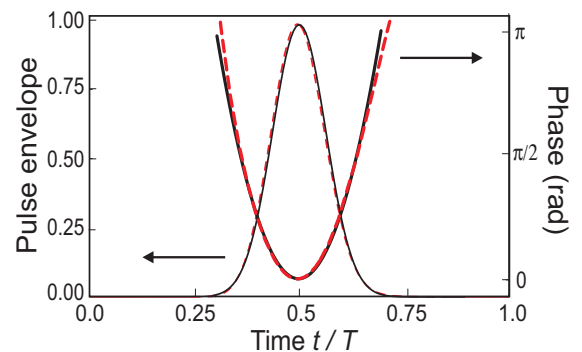


FIG. 4. (Color online) Envelope and phase of the input probe pulse (solid lines) and those of the first FID impulse (dashed red lines). The FID impulse was normalized to unity for a better comparison with the input pulse. Differences between the phase of the two pulses is observed only on the wings of the envelope, where the amplitude is small. Here,  $\Omega_c=50\Omega_p$  and  $\alpha z=1.5$ .

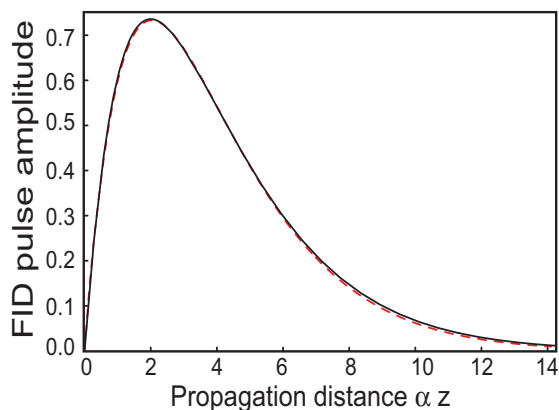


FIG. 5. (Color online) Peak amplitude of the first FID impulse shown in Fig. 3(a), evaluated at  $t=2.5T$ , as a function of propagation distance from numerically solving the Maxwell-Bloch equations (1) and (2) (dashed red line) and from the analytic solution of Eq. (12) (solid line).

quadratic phase  $\varphi(t)=0.6\pi(t/a)^2$  and tuned in between two resonances. Figure 4 shows the envelope and phase for the input pulse (solid line) and the first FID impulse (dashed line). Both have the same phase, with some minor discrepancies on the wings of the pulse envelope. And Fig. 5 shows the peak amplitude of the first FID impulse as a function of propagation distance. The agreement with Eq. (12) is excellent.

Coherent control of the moment of emission can be achieved by applying a sequence of coupling pulses. Figure 6 shows the total field observed when four coupling pulses are applied successively, each separated in time by a beat period  $T$ . In this case, the first FID impulse occurs only at a much later time. But, here, the FID impulse is  $\pi$  rad out of phase with the input pulse, as opposed to being in phase as that of Fig. 3(a). Each coupling pulse adds a  $\pi$  phase to the FID impulse; adding or removing one coupling pulse to the sequence would give a FID impulse in phase with the input pulse.

### VI. CONCLUSIONS

Previous studies on the coherent control of spontaneous emission are here extended into the ultrashort-pulsed regime. I studied the coherent control and suppression of the free-induction-decay emission from an excited multilevel atom. Typically, this emission is in the femto to picosecond time scale and so is the control pulse duration.

Suppression is achieved by means of a control pulse that connects the excited states to a lower state other than the ground state. This control pulse adjusts the phase of the ex-

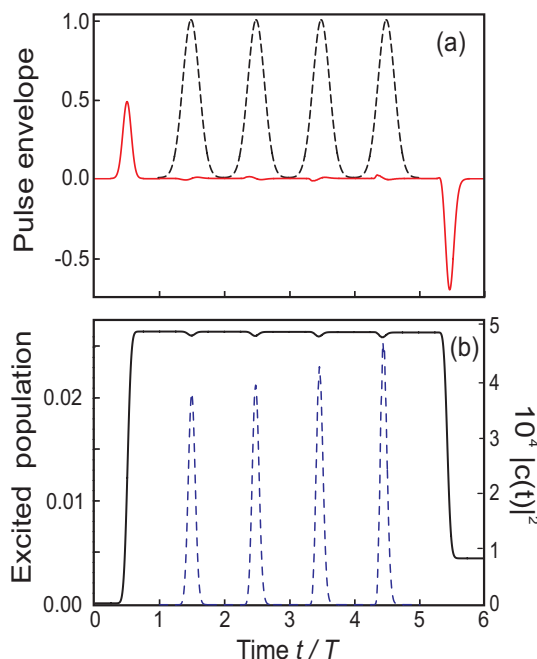


FIG. 6. (Color online) (a) The envelope of the probe pulse (solid red line), at  $\alpha z=1.5$ , in the presence of a sequence of four identical coupling pulses equally separated in time by the beat period  $T$  (dashed line). (b) Total population in the excited states (solid line) and in the lower state  $|2\rangle$  (dashed blue line).

cited states' probability amplitudes, inducing a destructive interference in the total atomic polarization. It is shown that the suppression is only temporary, and the free-induction decay reappears after one beat period of the excited manifold of states. Coherent control of the moment of emission is demonstrated by a sequence of equally spaced control pulses. As recently shown by Jacquy and colleagues [21], such a train of ultrashort pulses, with time delays on the order of hundreds of femtoseconds, can be easily implemented with a Fabry-Pérot interferometer.

It should be possible to extend this emission coherent control idea to weakly anharmonic systems as well. However, due to the anharmonicity of the system, the suppressed FID impulse that will appear one period later will not be temporally identical to the probe pulse, but it should be broader. If postponed for several beat periods by a sequence of control pulses, the FID impulse may come out significantly different from the probe pulse.

### ACKNOWLEDGMENTS

The author acknowledges the financial support of CNPq and FAEPEX.

- [1] S. E. Harris, *Phys. Today* **50**(7), 36 (1997).
- [2] A. M. Akulshin, S. Barreiro, and A. Lezama, *Phys. Rev. A* **57**, 2996 (1998).
- [3] S. Y. Zhu and M. O. Scully, *Phys. Rev. Lett.* **76**, 388 (1996).
- [4] E. Paspalakis and P. L. Knight, *Phys. Rev. Lett.* **81**, 293 (1998).
- [5] E. Paspalakis, C. H. Keitel, and P. L. Knight, *Phys. Rev. A* **58**, 4868 (1998).
- [6] M. A. G. Martinez *et al.*, *Phys. Rev. A* **55**, 4483 (1997).
- [7] J. Evers and C. H. Keitel, *Phys. Rev. Lett.* **89**, 163601 (2002).
- [8] Z. Ficek and S. Swain, *J. Mod. Opt.* **49**, 3 (2002).
- [9] G. S. Agarwal, M. O. Scully, and H. Walther, *Phys. Rev. Lett.* **86**, 4271 (2001).
- [10] E. Frishman and M. Shapiro, *Phys. Rev. Lett.* **87**, 253001 (2001).
- [11] T. Quang, M. Woldeyohannes, S. John, and G. S. Agarwal, *Phys. Rev. Lett.* **79**, 5238 (1997).
- [12] K. J. Vahala, *Nature (London)* **424**, 839 (2003).
- [13] M. Macovei and C. H. Keitel, *Phys. Rev. Lett.* **91**, 123601 (2003).
- [14] J. Arlt, C. Weiss, G. Torosyan, and R. Beigang, *Phys. Rev. Lett.* **79**, 4774 (1997).
- [15] J. N. Sweetser and I. A. Walmsley, *J. Opt. Soc. Am. B* **13**, 601 (1996).
- [16] I. P. Christov, *Opt. Commun.* **113**, 530 (1995).
- [17] L. E. E. de Araujo, *Phys. Rev. A* **72**, 053802 (2005).
- [18] L. E. E. de Araujo, *Phys. Rev. A* **73**, 053821 (2006).
- [19] R. de Vivie-Riedle *et al.*, *J. Phys. Chem.* **100**, 7789 (1996).
- [20] P. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953).
- [21] M. Jacquey, S. Bonhommeau, and M. A. Bouchene, *Opt. Lett.* **28**, 1272 (2003).