Ultrashort pulse propagation in multilevel systems

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Here, I discuss the propagation of an ultrashort pulse through a collection of harmonic multilevel systems. In the limit of weak excitation and a large number of excited states, I show that the amplitude of the input driving pulse decays exponentially with propagation distance. The absorption coefficient associated with this decay is determined by the characteristic time of the manifold of excited states, instead of the polarization decay time as in the conventional absorption coefficient of a two-level atom. The input ultrashort pulse creates in the excited states a wave packet, which oscillates emitting secondary pulses in the process. Analytic solutions are obtained that describe the propagation of individual wave-packet re-emission pulses, and it is shown that their phase depends on the detuning of the input pulse.

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The propagation of a weak ultrashort pulse in a two-level atom is well understood [1–4]. If the pulse spectrum is much wider than the resonance line (or equivalently, the pulse duration is much shorter than the coherence or polarization decay time T_2), then absorption and dispersion converts the pulse envelope into an oscillatory function of time. Since only a very small portion of the pulse spectrum is absorbed in the excitation process, the pulse can propagate many absorption lengths into the medium, thus deviating from Beer's law absorption. The propagation is properly described by linear dispersion theory, and the pulse shape can be calculated explicitly. The results of such a linear propagation model have been verified experimentally [5–7].

When more than two levels are considered, the dispersion properties of the medium will be changed significantly if the pulse spectrum is wide enough to overlap many excited states. Here, many frequencies are absorbed, and eventually removed, from the pulse spectrum by the multilevel medium during propagation. As a result, a train of secondary pulses is generated. From an alternate point of view, the ultrashort pulse creates a nonstationary superposition, or wave packet, in the multilevel medium. This wave packet oscillates with a characteristic time that corresponds to the quantum beat period between the excited states. These oscillations reflect themselves in the shape of the propagating ultrashort pulse: As it oscillates, the wave packet emits secondary pulses separated in time by that beat period, thus modifying the shape of the input driving pulse. The connection between the dynamics of a Rydberg wave packet and the shape of a propagating ultrashort pulse was established by Arlt and coworkers [8], along with their experimental observation of the reshaping of femtosecond laser pulses in Rydberg atoms of strontium. A significant deviation from Beer's law and the conventional area theorem for an ultrashort pulse propagating through a collection of multilevel systems was demonstrated by Sweetser and Walmsley [9]; they also observed experimentally the reshaping of ultrashort pulses in molecular systems. And Christov [10] has analyzed the inversionless amplification and the reshaping of ultrashort pulses in multilevel media.

In this paper, I study the propagation of an ultrashort pulse through a collection of multilevel atomic systems. I derive a propagation equation that, in principle, can be solved analytically for any of the secondary wave-packet pulses and provide analytic solutions for a few of them. These analytic solutions contain the explicit dependence of the wave-packet pulse amplitude on the propagation distance.

The model atomic system considered here is shown in Fig. 1. It consists of a ground state and a manifold of excited states. Many quantum systems, such as Rydberg atoms and diatomic molecules, have at least part of their energy structure of this type. Typically, these systems exhibit some degree of anharmonicity in their level structure. However, for simplicity, the model system considered here is of the harmonic type; this simplification allows one to obtain an analytic solution to the Maxwell–Bloch propagation equations. Nevertheless, the conclusions obtained for the harmonic system should be extendable to a mildly anharmonic system, at least qualitatively.

The ground state $|1\rangle$ is connected to each level $|n\rangle$ within the excited manifold by a dipole transition, but no direct

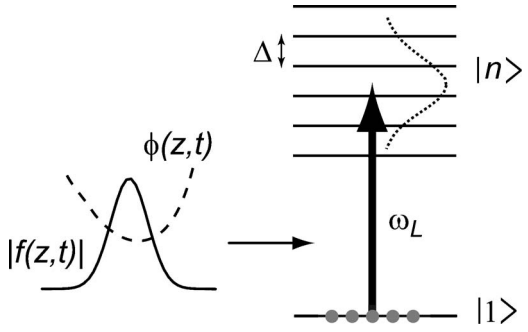

FIG. 1. The multilevel harmonic system, with level spacing Δ, is excited by a weak ultrashort pulse with amplitude $|f(z,t)|$, phase $\phi(z,t)$, and carrier frequency ω_L. The excited manifold contains a large number N of states, and the spectrum of the ultrashort pulse (dotted line) is wide enough to overlap many of these states. Solid circles represent the initial population distribution.

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interaction occurs between different levels of the excited manifold. When an ultrashort pulse is applied to the ground-state atom, many of the excited states within the manifold are coherently excited, and a wave packet is created. If Δ is the level spacing, the motion of this wave packet has a characteristic time of $T=2\pi/\Delta$. For an electronic Rydberg wave packet, T corresponds to the Kepler period [11,12], and for a vibrational wave packet in a diatomic molecule, it corresponds to the vibrational period [13], for example.

This interaction of the multilevel atom with a classical electric field $E(z,t)$ is properly modeled by the Hamiltonian

$$\hat{H}=\sum_{n=2}^{N}(\hbar\omega_n)|n\rangle\langle n|-E(z,t)\sum_{n=2}^{N}[d_n(|1\rangle\langle n|+|n\rangle\langle 1|)]. \quad (1)$$

The first term in the right-hand side of Eq. (1) corresponds to the unperturbed atomic Hamiltonian; the second term represents the dipole interaction between the atom and the electric field $E(z,t)$. Here, ω_n is the eigenfrequency of level n, with $\omega_1\equiv 0$; d_n is the electric dipole moment for the transition between the ground state and the excited state n; and N is the number of states in the excited manifold. In this model, dipole-dipole interactions between pairs of atoms are neglected.

Anywhere in the medium, the electric field is written as $E(z,t)=E_0 f(z,t)\exp[-i(kz-\omega_L t)]+c.c.$ The dimensionless envelope function $f(z,t)$ is complex: $f(z,t)\equiv|f(z,t)|\exp[i\phi(z,t)]$. The modulus $|f(z,t)|$ is thus the field's slowly varying amplitude, $\phi(z,t)$ is its phase (also slow varying), and ω_L is its center frequency. At the input ($z=0$), the driving field's envelope $|f(z,t)|$ has a maximum amplitude equal to one. The input pulses to be considered here have durations that are shorter than the characteristic time T of the multilevel system. For a Rydberg atom, $T\lesssim 100$ ps, and for a diatomic molecule such as K_2, $T\approx 500$ fs. Therefore, the input pulses are many orders of magnitudes shorter than the polarization decay time T_2 or any other relaxation time of the system. This is why the Hamiltonian of Eq. (1) contains no relaxation terms, such as spontaneous decay, for example.

The state of the system is expanded in terms of the unperturbed eigenstates of the atom:

$$|\Psi(z,t)\rangle=a(z,t)|1\rangle+\sum_{n=2}^{N}b_n(z,t)e^{-i\omega_n t}|n\rangle, \quad (2)$$

where $a(z,t)$ is the probability amplitude of the ground state $|1\rangle$, and $b_n(z,t)$ is the probability amplitude of the various excited states $|n\rangle$.

Equations (1) and (2) are next substituted into Schrödinger's equation. In the rotating wave approximation, the temporal dynamics of the probability amplitudes is described by

$$\dot{a}(z,t)=0.5\,i\,\Omega f^{*}(z,t)\sum_{n=2}^{N}b_n(z,t)e^{-i\delta_n t}, \quad (3a)$$

$$\dot{b}_n(z,t)=0.5\,i\,\Omega f(z,t)a(z,t)e^{i\delta_n t}, \quad (3b)$$

The following quantities have been defined: $\Omega\equiv 2dE_0/\hbar$ is the Rabi frequency and $\delta_n\equiv\omega_n-\omega_L$ is the laser frequency detuning for each bound-excited state transition. The electric-dipole moments for transitions between the ground and excited states have been assumed to be approximately constant over the entire excited-state manifold ($d_n\approx d$). And the atom starts with all the population initially in its ground electronic state; that is, $a(z,0)=1$ and $b_n(z,0)=0$.

The medium is optically thick, and the spatial evolution of the ultrashort pulse in such a medium is described by the reduced wave equation [9]:

$$\frac{\partial}{\partial z}[\Omega f(z,\tau)]=2i\mu\sum_{n=2}^{N}P_n(z,\tau), \quad (4)$$

where $P_n(z,\tau)=[b_n(z,\tau)\exp(-i\delta_n\tau)]a^{*}(z,\tau)$ is the induced polarization in the medium; $\mu=\omega_L\mathcal{N}d^2/\epsilon_0\hbar c$ is the coupling constant between the field and the polarization; $\mathcal{N}$ is the number density; ϵ_0 is the permittivity of the multilevel medium for nonresonant transitions; and $\tau=t-z/v_g$, with v_g being the group velocity of the pulse at the center of the spectrum. The Maxwell-Bloch Eqs.(3), with t replaced by the local time τ, and Eq. (4) must be solved together.

In the weak excitation regime, where $a(z,\tau)\approx 1$, the probability amplitudes of the excited states are, from Eq. (3b): $b_n(z,\tau)\approx 0.5\,i\Omega\int_{-\infty}^{\tau}ds f(z,s)\exp(i\delta_n s)$. Substituting this result into the wave equation yields:

$$\frac{\partial}{\partial z}f(z,\tau)=-\mu\int_{-\infty}^{\tau}ds f(z,s)\sum_{n}e^{i\delta_n(s-\tau)}. \quad (5)$$

Since the levels are equally spaced, I can write $\delta_n=\bar{\Delta}+\Delta(n-\bar{n})$, where $\bar{\Delta}=\omega_{\bar{n}}-\omega_L$ is the detuning of level $\bar{n}$, the average quantum number of the states excited by the driving field. Substituting $m=n-\bar{n}$ gives

$$\sum_{n}e^{-i\delta_n(\tau-s)}\approx e^{-i\bar{\Delta}(\tau-s)}\sum_{m=-\infty}^{\infty}e^{-im\Delta(\tau-s)}=\left(\frac{2\pi}{\Delta}\right)e^{-i\bar{\Delta}(\tau-s)}\sum_{m=-\infty}^{\infty}\delta\left(\tau-s-\frac{2\pi}{\Delta}m\right). \quad (6)$$

Since the exponentials will cut off the contributions of levels with large detunings, the limits in the sum can be extended to infinity to a good aproximation. The equality comes from the Poisson sum formula [14]. Substituting this approximation back into Eq. (5) and performing the integration yields

$$\frac{\partial}{\partial z}f(z,\tau)=-\alpha\left[\frac{1}{2}f(z,\tau)+\sum_{m=1}^{\infty}f(z,\tau-2\pi m/\Delta)e^{-im\theta}\right], \quad (7)$$

where

$$\alpha = 2\pi\mu/\Delta = \omega_L \mathcal{N} d^2 T/\epsilon_0 \hbar c, \tag{8}$$

and $\theta \equiv 2\pi\bar{\Delta}/\Delta$. The negative-$m$ terms in the summation have been dropped since $s \leqslant \tau$ in Eq. (5). According to Eq. (7), the present spatial evolution of the electric field depends on sampled past impulses. This sampling occurs at discrete times that are integer multiples of the characteristic time $T = 2\pi/\Delta$. Each of these sampled impulses corresponds either to the input driving pulse or to a secondary pulse radiated by the induced polarization in the multilevel medium—a wave packet recurrence.

For times less than the characteristic time ($0 \leqslant \tau < T$), the wave equation becomes

$$\frac{\partial}{\partial z} f(z,\tau) = -\frac{1}{2}\alpha f(z,\tau), \tag{9}$$

which can be easily solved, giving:

$$f(z,\tau) = f(0,\tau) e^{-\alpha z/2}. \tag{10}$$

Therefore, the input ultrashort driving impulse propagates with its initial form preserved, except for the multiplicative Beer's law decay factor $\exp(-\alpha z/2)$. The conventional absorption coefficient α entering Beer's law is affected by the polarization decay time T_2 [2]. Here, however, due to the *quasi*-impulsive nature of the excitation, T_2 is many orders of magnitude larger than the incident pulse width, and it does not affect α. As a matter of fact, as pointed out by Sweetser and Walmsley [9], T_2 can be made to be arbitrarily large without affecting the shape of the transmitted ultrashort pulse, leading to a deceivingly large conventional optical density $\alpha z = \mu z T_2$. For this reason, they prefer to use μz, which has units of inverse time, to characterize a multilevel medium. From Eq. (8), α is determined by the much shorter characteristic time T of the excited manifold. Thus, it is more appropriate and natural to characterize the multilevel medium by the much shorter, and unitless, optical density $\mu z T$. Likewise, the absorption length $\zeta = 1/\alpha$ of such a multilevel medium will be much longer than that of the conventional two-level medium with T_2 as its characteristic time. Considering the experimental conditions for the X→ A transition in potassium dimers [15]: $\mathcal{N} = 5 \times 10^{14} \mathrm{cm}^{-3}$, $d = 11.4$ D, $\lambda_L = 2\pi c/\omega_L = 830$ nm, and $T \approx 500$ fs (the vibrational period of the A state), then from Eq. (8), the absorption length for this system is $\zeta \approx 4$ mm.

For the next time interval ($T \leqslant \tau < 2T$ and $m = 1$),

$$\frac{\partial}{\partial z} f(z,\tau) = -\frac{1}{2}\alpha f(z,\tau) - \alpha e^{-i\theta} f(z,\tau - T), \tag{11}$$

where $f(z,\tau - T)$ is the solution from the previous interval, given by Eq. (10). One then finds:

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

where $f(0,\tau - T)$ is the input driving field shifted in time. The impulse of Eq. (12), which corresponds to the first wave-packet recurrence, has an identical temporal shape to that of the driving field; their relative phase θ is set by the detuning of the driving field. If the laser's center frequency is set exactly to one of the atomic resonances, then $\theta = 0$ and the wave packet recurrence is π radians out of phase with the driving field. If the laser is tuned half way between two resonances ($\theta = \pi$), the two impulses are in phase.

TABLE I. Analytic solutions for the driving field ($m = 0$) and the first four wave-packet recurrences.

m	$f(z,\tau)$
0	$e^{-\alpha z/2} f(0,\tau)$
1	$-(\alpha z) e^{-\alpha z/2} e^{-i\theta} f(0,\tau - T)$
2	$\left[\frac{1}{2}(\alpha z)^2 - \alpha z\right] e^{-\alpha z/2} e^{-2i\theta} f(0,\tau - 2T)$
3	$\left[-\frac{1}{6}(\alpha z)^3 + (\alpha z)^2 - \alpha z\right] e^{-\alpha z/2} e^{-3i\theta} f(0,\tau - 3T)$
4	$\left[\frac{1}{24}(\alpha z)^4 - \frac{1}{2}(\alpha z)^3 + \frac{3}{2}(\alpha z)^2 - \alpha z\right] e^{-\alpha z/2} e^{-4i\theta} \times f(0,\tau - 4T)$

In principle, Eq. (7) could be solved for any of the wave-packet recurrences, as long as these recurrences occur on time scales much shorter than the polarization decay time. At times mT comparable to T_2, the Hamiltonian of Eq. (1) will no longer be valid. Table I summarizes the solutions for the first four recurrences ($m = 4$). Although each of the impulses contains an exponential damping factor, only the driving impulse ($m = 0$) follows a pure Beer's law decay. The exponential decay is accompanied by an "extra" polynomial z-dependence indicating that, given a long enough propagation distance, every impulse eventually will be completely absorbed by the medium. As suggested by the results presented in Table I, every impulse is identical (in time) to the input pulse, and their relative phase depends on its detuning. Furthermore, given these results it is reasonable to expect that the area of the total electric field within the medium, $\int_{-\infty}^{\infty} \Sigma_m \Omega f(z,\tau - mT) d\tau$, should deviate significantly from Beer's Law, as has been previously verified [9].

I tested the validity of the analytic solutions shown in Table I by comparing them with a numerical solution of the Maxwell–Bloch Eqs. (3) and (4) for two test driving pulses.

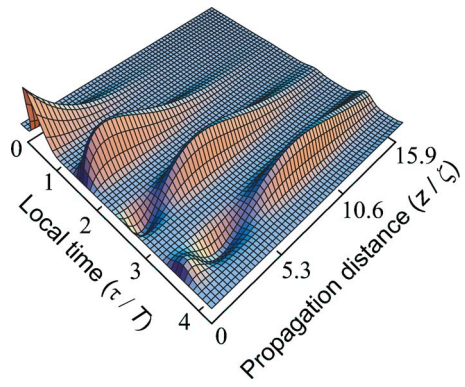

FIG. 2. (Color online) The amplitude of the electric field $f(z,\tau)$ as a result of propagation inside the multilevel medium. The input pulse is a Gaussian impulse, given by Eq. (13), at $z = 0$. Time is measured in units of the characteristic time $T = 2\pi/\Delta$, and distance is measured in units of the absorption length ζ.

The first test pulse consisted of a transform-limited Gaussian pulse given by

$$f(0,\tau) = \exp[-(4\ln 2)(\tau-\tau_0)^2/a^2], \quad (13)$$

where a is its full width at half maximum, and $\tau_0=0.5T$. With the pulse width set to $a=0.3T$, the spectrum of the driving pulse overlaps about seven states in the excited manifold. The pulse is tuned exactly half way between two resonances ($\theta=0$). The amplitude E_0 was set so that the pulse area was $\Omega\int_{-\infty}^{\infty} f(0,\tau)d\tau=0.01\pi$. (Under these conditions, a numerical calculation shows that the driving pulse transfers a very small fraction, approximately 0.05%, of the ground-state population to the excited states.)

Propagation of this test pulse in a multilevel medium containing $N=12$ excited states is shown in Fig. 2. Four impulses are observed in the figure: The input driving pulse and three wave-packet recurrences. The driving pulse and the first wave-packet recurrence are in phase as predicted by Eq. (12). The amplitude of the input pulse decays monotonically, but those of the other impulses have a more complicated z dependence, even changing signs as the impulse propagates into the medium. The first wave-packet recurrence, for example, is initially amplified up to a propagation distance of about $z=2.5\zeta$ when it begins to be absorbed by the medium, and its amplitude starts decaying. Figure 3 compares the numerically obtained z dependence of each impulse with that of the analytic solutions of Table I. As can be seen, extending the limits in the sum in Eq. (6) to infinity is indeed a good approximation: The analytic solutions reproduce very well the numerical results, at least for the first few wave-packet recurrences. However, the higher the recurrence number m, the worst the agreement should be between the two solutions. This is due to the small finite number of excited states N considered in the numerical calculation. Increasing N should improve this agreement.

Figure 4 shows the numerically propagated test pulse when it is tuned half way ($\theta=\pi$) between two excited states and on resonance ($\theta=0$) with one of the atomic transitions. Here, $z=0.2\ \zeta$. At this propagation distance, all the secondary pulses shown are in phase with each other and π radians out of phase with the input pulse, when in resonance. When the excitation laser is tuned half way between two transitions, only the odd-numbered secondary pulses are in phase with the excitation laser. These observations agree with the results presented in Table I.

The prediction that all wave-packet recurrences should maintain during propagation a temporal amplitude and phase identical to those of the input pulse was checked with a second test pulse. This test pulse was similar to the first one, except for a cubic phase:

$$f(0,\tau) = \exp[-(4\ln 2)(\tau-\tau_0)^2/a^2 + i\varphi(\tau-\tau_0)], \quad (14)$$

where $\varphi(\tau)=0.42\pi\tau^3$.

Figure 5 shows the predicted $m=1$ impulse from Table I along with the corresponding numerically calculated impulse after propagating a distance $z=2.5\ \zeta$. At this propagation distance, the impulse is close to its maximum amplitude. The agreement between the two is excellent with respect to both amplitude and phase (discrepancies occur where the amplitude is negligible).

In summary, I have studied the propagation of a weak ultrashort pulse in a harmonic multilevel medium and have calculated explicit analytic expressions for the shape of various of the secondary impulses (wave packet re-emissions) that constitute the total electric field inside the medium.

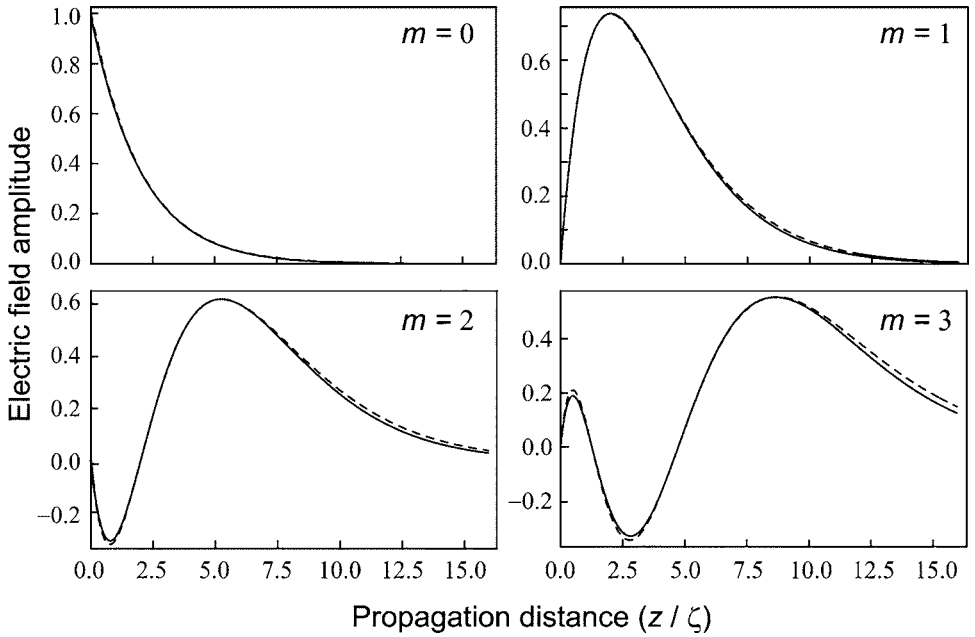

FIG. 3. Spatial dependence of the input pulse ($m=0$) and the first three wave-packet recurrences ($m=1$, 2, and 3) for both numerical (solid line) and analytic (dashed line) solutions. The spatial dependences are evaluated at the center of the temporal profile of each of the impulses: $\tau=\tau_0+mT$.

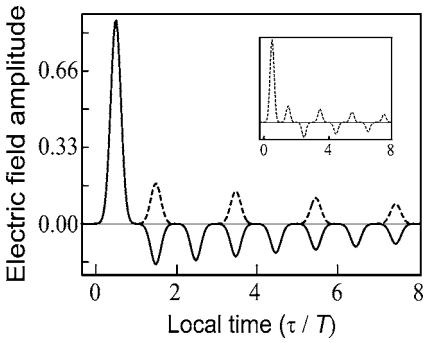

FIG. 4. The numerically propagated test pulse of Eq. (13) for two different detunings: the input pulse tuned to a resonance (solid line) and tuned in between two levels (dashed line). The inset shows the latter case separately for better visualization.

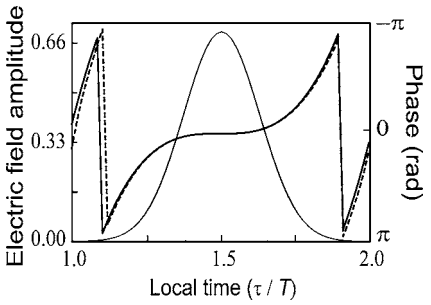

FIG. 5. The amplitude and phase of the first wave-packet recurrence (dashed line) and its corresponding analytic solution from Table I (solid line) at a propagation distance of $z=2.5\,\zeta$. The input pulse (at $z=0$) is given by Eq. (14), and it corresponds to a Gaussian impulse with a cubic phase.

These analytic solutions were obtained under the approximation that the excited manifold contains a large number of states, although only some of them are populated. I have shown that the amplitude of the propagating input pulse follows a Beer's law decay, where the absorption coefficient is determined by the characteristic time of the excited manifold. Furthermore, each of the secondary impulses, anywhere inside the medium, is an exact temporal copy of the input pulse, and their phase relative to the driving field is set by its detuning. The insights developed here should also apply to media, such as diatomic molecules, which exhibit some mild degree of anharmonicity; this should be true at least to the first few vibrational periods when this anharmonicity has not manifested itself too strongly yet [13].

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