Few-cycle pulses in two-level media

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Techniques for producing, measuring, and applying ever shorter electromagnetic pulses are being developed for incorporation in a variety of modern high-speed systems. In many cases these pulses are at most a few cycles in length, and so-called half-cycle electromagnetic pulses are also widely employed. The interaction of such pulses with two-level media is considered here in detail, and these media are basic to many of the absorbing and amplifying configurations of optics and laser studies. Significant delays and distortion of the resulting polarization and population pulses can occur, and nonlinear optical effects are also revealed. The limitations of the parity, rate-equation, and rotating-wave approximations for the characterization of such few-cycle interactions are also explored. [S1050-2947(98)03201-6]

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I. INTRODUCTION

Some of the most basic problems in physics involve the interaction of electromagnetic fields with atoms or molecules. One fundamental starting point for such studies employs the Maxwell-Heaviside equations for the electromagnetic fields in combination with the Schrödinger or Dirac equation for the atoms. For the high-frequency fields associated with transitions between atomic levels, it is usually possible to treat the fields as harmonic in time and space, with an envelope that may vary slowly in time compared to an optical cycle or slowly in space compared to a wavelength. In these cases it has generally been possible to employ the rotating-wave approximation in the atom equations and the slowly-varying-envelope approximations in the field equations. Sometimes the rate-equation approximation is also applicable. These approximations dramatically simplify most calculations of practical relevance. Recently, however, there has been increasing interest in a class of field variations that does not fit so conveniently with these established approximations.

One of the persistent trends in laser studies has been the development of systems capable of producing ever shorter optical pulses. Thus it has been possible, with colliding-pulse mode-locked dye lasers, to directly obtain pulses that are as short as 27 fs in duration at a wavelength of about 630 nm [1]. Using pulse compression external to a colliding-pulse mode-locked dye laser cavity, it has been possible to shorten pulses centered at about 620 nm to a length of only about 6 fs [2]. With the invention of the self-mode-locked titanium: sapphire laser [3], it became possible to obtain pulses shorter than 10 fs without external pulse compression. Pulses of about 7.5 fs length have now been obtained directly from these lasers at a wavelength of about 800 nm [4]. For still shorter pulses, fiber-optic pulse compressors are useful, and have led to the generation of pulses that are about 4.5 fs in duration at 780 nm [5,6]. For direct titanium:sapphire laser systems, it is anticipated that a further shortening by about a factor of 2 will be possible with the development of improved dispersion-compensation schemes [7], and improvements in the pulse compression results may also be expected.

Techniques for producing, measuring, and applying ultrashort electromagnetic pulses are being developed for incorporation in a variety of modern high-speed systems. As an example of a fundamental application, short pulses are sometimes used to manipulate atomic wave functions in testing the predictions of quantum mechanics [8]. At a more practical level, fs pulses such as those described above have been employed in the measurement and characterization of the electronic properties of materials and devices [9]. However, at the optical frequencies used, these pulses are sometimes only a few cycles in length; and it is not always clear how to interpret experimental data. Thus the period of a sinusoidal wave is related to its wavelength by the formula t_0 (fs)$\approx\frac{10}{3}\lambda$ (μm), and this is 2.6 fs for a wavelength of 780 nm. Comparing this result to the recent pulse-length data summarized above, one finds that the shortest optical pulses are only about two or three cycles in length. In such cases the fields cannot readily be described as almost sinusoidal, and the validity of standard approximations may be in doubt. The slowly-varying-envelope derivative approximations, for example, require that the bandwidth associated with a transition be small compared to the optical frequency. This condition is not well satisfied for wide-band dye and titanium:sapphire lasers, and one should expect significant discrepancies between experimental results and the approximate theories for several amplifier [10,11] and oscillator [12] configurations. The basic rotating-wave approximation would also fail in such systems [13]. Even the typical nonlinear techniques employed to measure the pulse length may not always behave as one would expect. Looked at more optimistically, there may in these cases be new physical effects which would have been overlooked in conventional analyses.

Once a source for optical pulses of a few fs length has been developed, nonlinear techniques can be employed to obtain pulses of similar length but having either a higher or lower carrier frequency. Clearly, the carrier frequency does not have to be lowered far below the optical range before the resulting pulses would appear to be less than one cycle

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in length. Thus, a 5-fs pulse at a wavelength of 1.5 μm would be about one cycle in duration, and at a wavelength of 3.0 μm would qualify as a so-called half-cycle pulse with no significant field reversals during the pulse period. In such cases some of the standard approximations have no validity at all, and more fundamental methods are required for the treatment of the interaction of fields with atoms. One of the most popular examples of such pulse down-shifting employs few-fs pulses for the transient introduction of free carriers into a biased semiconductor. The resulting current pulse can radiate sub-single-cycle THz/or mm wave pulses [14]. Such pulses have recently found many applications, particularly again in the manipulation of atomic wave functions [15].

An emphasis in this study is on the resonant and near-resonant interaction of fs pulses with two-level media. To the extent that an interaction involves only a single transition, a real atom may be approximated as having only two levels, and this approximation has been widely employed in interpreting and predicting observable phenomena [16–18]. However, it is important to remark at least briefly on possible limitations of the two-level model for representing short-pulse electromagnetic interactions with practical media. While there may be some intrinsic mathematical interest in models like those to be developed here, we would also like to believe that these models can correspond at least approximately to actual physical systems. Thus, for example, it might be possible to find a system with two electromagnetically coupled energy levels that are far removed from all other states, but in practice energy levels are often more-or-less uniformly distributed. Also, the spectrum associated with very short pulses may have a width that is comparable to the underlying carrier frequency. For such cases one can ask whether two-level models might still sometimes be realistic.

There are some specific circumstances that are to be avoided if one wishes to use a two-level approximation in a multilevel system. First, it is important that all of the frequency content of the input pulse be close to (or less than if there are no intermediate states) the energy spacing between the two levels in the model. If this is not the case, then it is possible that other levels will also be interacting with the field. In most of the examples given here the carrier frequency is at or below the transition frequency, and the spectral broadening due to the pulse envelope is smaller than the carrier frequency. In this respect, then, these examples are not inconsistent with the idea of a two-level model.

Another likely constraint on use of a two-level model is that the field amplitude must not be too large. In a system approximated as a two-level absorber, for example, absorption of the field brings the atoms or molecules from the ground state into the upper state. These excited-state atoms might then be available for secondary excitation by some coherent or incoherent process to other higher-lying levels, or to ionization. The efficiency of such processes depends on the amplitude of the field, and would be a serious concern if a significant fraction of the atoms in the model were brought into the upper state and remained there in the continuing presence of the field. However, in most of our examples the pulse field will be kept weak enough that only at most about 20% of the population is brought to the upper level. While these arguments can only be qualitative, it does seem that, as others have concluded, there may be significant regions of practical parameter space where the two-level model would be sufficient for the treatment of short-pulse interactions.

In addition to the two-level assumption, most previous treatments employ further approximations in calculating the response of an atomic medium to an applied field. The validity of these approximations is not always assured when the field is in the form of a pulse only a few cycles in duration. For example, one almost universally employed simplification is known as the rotating-wave approximation, and the usual reason for employing this approximation is to achieve greater analytical simplicity. The resulting models have more slowly evolving variables, and the solution methods are always more straightforward. Thus it is a matter of considerable practical importance to know the conditions under which the rotating-wave approximation may be sufficiently accurate for a particular application.

Recently, some effects of few-cycle electromagnetic pulses on the populations of two-level absorbing systems have been considered [19,20]. Here we will be treating general dipole moment configurations, and we will establish a framework for the description of two-level media with arbitrary inhomogeneous broadening. In addition to investigating the evolution of the level populations, it will be of interest to observe the development of the oscillating polarization during and after the short excitation pulse, and the validity and consequences of the rotating-wave-approximation for the characterization of few-cycle interactions is explored. Inconsistent with that approximation, we find that there can be significant delays and distortion of the polarization and population pulses that result from such interactions, and nonlinear optical effects are found. Other approximations examined include the parity approximation and the rate-equation approximation.

A general semiclassical model is briefly developed in Sec. II for the dynamics of a laser medium having arbitrary levels of homogeneous and inhomogeneous line broadening. Use of this starting point provides a common basis for this analysis and previous investigations of laser instabilities [21], space [10,11] and time [12] derivative approximations, and the rotating-wave approximation [13]. The reduction of the general model to a simpler and more specific set of equations for a homogeneously broadened medium interacting with few-cycle pulses is discussed in Sec. III. Numerical solutions of the model are described in Sec. IV, and the delays and distortions of the polarization response in comparison to the applied field are discussed. The limitations of the parity approximation in short-pulse systems is also discussed in Sec. IV. Under some conditions a rate-equation-like approximation may be applicable even when the rotating-wave approximation is not, and this situation is discussed in Sec. V. The rotating-wave approximation is generally not valid for few-cycle pulses, as considered in Sec. VI, and the physical effects of several parameter variations are briefly treated in Sec. VII.

II. GENERAL MODEL

In investigating the interaction of very short pulses with material systems, it is necessary at an early stage to restrict

the classes of interaction to be considered. The reason for such a restriction is, of course, the excessively vast and diverse array of possible physical effects that one would need to include for a truly complete model. As suggested by the title, our main emphasis will be on interactions with media possessing two distinct energy states. When the frequency of incident radiation is near resonance with a transition in a localized ensemble of atoms or molecules, absorptions or emissions may be induced between the corresponding states. The resulting redistribution of population can in turn change the dipole moment of the ensemble, and this time-dependent dipole moment can contribute to the overall electromagnetic field in the region of interest. On a larger scale this effect can lead to a time dependence of the macroscopic polarization, including changes in the index of refraction and the loss or gain. Behavior of this sort is well known in solids, liquids, and gases, and one purpose here will be to develop methods for treating such resonant interactions when the electromagnetic pulses are only a few cycles in length.

Our starting point for this semiclassical study will be the usual density-matrix equation [22]

$$\frac{\partial\tilde{\rho}}{\partial t}=-\frac{i}{\hbar}[\tilde{H},\tilde{\rho}], \tag{1}$$

where the right-hand side includes the commutator of the matrix form of the Hamiltonian operator H (between eigenfunctions of the unperturbed system) with the density matrix ρ. As mentioned above, near-resonant interactions can often be treated including only two strongly coupled energy states. Transitions involving other states are regarded as being so far from resonance with the incident field that the inclusion of simple phenomenological relaxation terms provides an adequate description of their effects. In this familiar case Eq. (1) represents four equations for the elements of the 2×2 density matrix, and these equations can be written in expanded form as

$$\frac{\partial\rho_{21}}{\partial t}=-\frac{i}{\hbar}[H_{21}\rho_{11}+H_{22}\rho_{21}-\rho_{21}H_{11}-\rho_{22}H_{21}], \tag{2}$$

$$\frac{\partial\rho_{22}}{\partial t}=-\frac{i}{\hbar}[H_{21}\rho_{12}-\rho_{21}H_{12}], \tag{3}$$

$$\frac{\partial\rho_{11}}{\partial t}=-\frac{i}{\hbar}[H_{12}\rho_{21}-\rho_{12}H_{21}], \tag{4}$$

$$\rho_{12}=\rho_{21}^{*}, \tag{5}$$

where the subscripts 2 and 1 refer, respectively, to the higher- and lower-energy states of the transition. Equation (5) is written using the fact that the density matrix is Hermitian.

The Hamiltonian operator will now be separated into a part H_0, which depends only on the static background fields experienced by an electron, and a part H', which represents the effects of the applied electromagnetic field. For the systems of interest here, the interaction with the applied field can be written in the form

$$H'=-\boldsymbol{\mu}\cdot\mathbf{E}, \tag{6}$$

where $\boldsymbol{\mu}=e\mathbf{r}$ is the dipole moment operator, and $\mathbf{E}$ is the electric field, assumed constant over the dimensions of the atom. With these substitutions Eqs. (2)–(4) become

$$\begin{aligned}\frac{\partial\rho_{21}}{\partial t}&=-\frac{i}{\hbar}[(H_0+H')_{21}\rho_{11}+(H_0+H')_{22}\rho_{21}\\&\quad-\rho_{21}(H_0+H')_{11}-\rho_{22}(H_0+H')_{21}]\\&=-\frac{i}{\hbar}[(E_2-E_1)\rho_{21}+\boldsymbol{\mu}_{21}\cdot\mathbf{E}(\rho_{22}-\rho_{11})\\&\quad-(\boldsymbol{\mu}_{22}-\boldsymbol{\mu}_{11})\cdot\mathbf{E}\rho_{21}],\end{aligned} \tag{7}$$

$$\begin{aligned}\frac{\partial\rho_{22}}{\partial t}&=-\frac{i}{\hbar}[(H_0+H')_{21}\rho_{12}-\rho_{21}(H_0+H')_{12}]\\&=-\frac{i}{\hbar}\mathbf{E}\cdot(\boldsymbol{\mu}_{12}\rho_{21}-\boldsymbol{\mu}_{21}\rho_{12}),\end{aligned} \tag{8}$$

$$\begin{aligned}\frac{\partial\rho_{11}}{\partial t}&=-\frac{i}{\hbar}[(H_0+H')_{12}\rho_{21}-\rho_{12}(H_0+H')_{21}]\\&=-\frac{i}{\hbar}\mathbf{E}\cdot(\boldsymbol{\mu}_{21}\rho_{12}-\boldsymbol{\mu}_{12}\rho_{21}).\end{aligned} \tag{9}$$

In these results E_2 and E_1 are the energy eigenvalues, and we have used the fact that the eigenfunctions of the background Hamiltonian H_0 are an orthogonal set. It is usual to replace the energy difference by its frequency equivalent $\hbar\omega_0$, where ω_0 is the center frequency of the transition. With this substitution, Eq. (7) can be written

$$\frac{\partial\rho_{21}}{\partial t}=-i\left[\omega_0+\frac{1}{\hbar}(\boldsymbol{\mu}_{22}-\boldsymbol{\mu}_{11})\cdot\mathbf{E}\right]\rho_{21}-\frac{i}{\hbar}\boldsymbol{\mu}_{21}\cdot\mathbf{E}(\rho_{22}-\rho_{11}). \tag{10}$$

Equations (5) and (8)–(10) describe the behavior of the density matrix in terms of the applied electromagnetic field. From the density matrix it is possible to derive the polarization of an ensemble of atoms. The dipole moment (or the ensemble average of the expectation value of the dipole moment operator) for an atom can be written

$$\mathbf{p}=\mathrm{tr}(\tilde{\rho}\tilde{\boldsymbol{\mu}})=\rho_{11}\boldsymbol{\mu}_{11}+\rho_{12}\boldsymbol{\mu}_{21}+\rho_{21}\boldsymbol{\mu}_{12}+\rho_{22}\boldsymbol{\mu}_{22}. \tag{11}$$

If this result for a single atom is integrated over a macroscopic ensemble of atoms, one obtains an expression for the polarization of the resonant medium. This polarization in turn will contribute to the behavior of the overall electromagnetic field.

As noted previously, focusing this semiclassical analysis on transitions between only two energy states would generally require some phenomenological method of incorporating transitions to and from other states of the system. A generalization to include inhomogeneous broadening might sometimes be useful as well. Thus, we rewrite Eqs. (5) and (8)–(10) in the more complete forms

$$\left(\frac{\partial}{\partial t}+v\,\frac{\partial}{\partial z}\right)\rho_{21}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$=-\left[i\omega_\alpha+\frac{i}{\hbar}\,(\boldsymbol{\mu}_{22}-\boldsymbol{\mu}_{11})\cdot\mathbf{E}(z,t)+\gamma\right]\rho_{21}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$-\frac{i}{\hbar}\,\boldsymbol{\mu}_{21}\cdot\mathbf{E}(z,t)[\rho_{22}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$-\rho_{11}(v,\omega_\alpha,\theta,\phi,z,t)], \tag{12}$$

$$\left(\frac{\partial}{\partial t}+v\,\frac{\partial}{\partial z}\right)\rho_{22}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$=\lambda_2(v,\omega_\alpha,\theta,\phi,z,t)-\gamma_2\rho_{22}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$-\frac{i}{\hbar}\,\mathbf{E}(z,t)\cdot[\boldsymbol{\mu}_{12}\rho_{21}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$-\boldsymbol{\mu}_{21}\rho_{12}(v,\omega_\alpha,\theta,\phi,z,t)], \tag{13}$$

$$\left(\frac{\partial}{\partial t}+v\,\frac{\partial}{\partial z}\right)\rho_{11}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$=\lambda_1(v,\omega_\alpha,\theta,\phi,z,t)-\gamma_1\rho_{11}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$+\gamma_{21}\rho_{22}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$+\frac{i}{\hbar}\,\mathbf{E}(z,t)\cdot[\boldsymbol{\mu}_{12}\rho_{21}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$-\boldsymbol{\mu}_{21}\rho_{12}(v,\omega_\alpha,\theta,\phi,z,t)], \tag{14}$$

$$\rho_{12}(v,\omega_\alpha,\theta,\phi,z,t)=\rho_{21}^{*}(v,\omega_\alpha,\theta,\phi,z,t)], \tag{15}$$

where γ_2 and γ_1 are the total decay rates of the upper and lower levels, respectively, γ_{21} is the rate of direct decays from level 2 to level 1, γ is the decay rate of the off-diagonal elements, λ_2 and λ_1 are the pumping rates, and the notation c.c. means the complex conjugate of the preceding terms. The laser medium is assumed to have both Doppler and non-Doppler inhomogeneous broadening mechanisms, with v being the z component of the velocity, and ω_α the center frequency of the laser transition for members of an atomic or molecular class α. The medium is also assumed to have an orientational distribution of transition moments, with the spherical coordinates θ and ϕ distinguishing the orientational classes.

To the density-matrix equations for the atomic or molecular populations and polarizations must be added an equation for the electric field. The wave equation for the electric field of a linearly polarized wave in a laser medium can be written

$$\frac{\partial^2\mathbf{E}(z,t)}{\partial z^2}-\mu_1\sigma\,\frac{\partial\mathbf{E}(z,t)}{\partial t}-\mu_1\varepsilon_1\,\frac{\partial^2\mathbf{E}(z,t)}{\partial t^2}=\mu_1\,\frac{\partial^2\mathbf{P}(z,t)}{\partial t^2}. \tag{16}$$

The permeability μ_1 and permittivity ε_1 should be understood to include all of the magnetic and dielectric properties of the laser medium except for the polarization $\mathbf{P}(z,t)$, which is due to the lasing atoms or molecules. From Eq. (11) the polarization driving this equation can be related back to the off-diagonal density matrix elements by

$$\mathbf{P}(z,t)=\int_0^{2\pi}\int_0^{\pi}\int_0^{\infty}\int_{-\infty}^{\infty}n(v,\omega_\alpha,\theta,\phi,z,t)$$

$$\times[\boldsymbol{\mu}_{11}\rho_{11}(v,\omega_\alpha,\theta,\phi,z,t)+\boldsymbol{\mu}_{21}\rho_{12}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$+\boldsymbol{\mu}_{12}\rho_{21}(v,\omega_\alpha,\theta,\phi,z,t)$$

$$+\boldsymbol{\mu}_{22}\rho_{22}(v,\omega_\alpha,\theta,\phi,z,t)]dv\;d\omega_\alpha d\Omega, \tag{17}$$

where $n(v,\omega_\alpha,\theta,\phi,z,t)dv\;d\omega_\alpha d\Omega$ is the number of molecules per unit volume at position z and time t having their z component of velocity between v and $v+dv$, their intrinsic transition frequency between ω_α and $\omega_\alpha+d\omega_\alpha$, and their orientation within the solid angle $d\Omega$ about the (θ,ϕ) direction. Equations (12)–(17) are a complete set from which the time and space dependences of the electric field and of the atomic or molecular parameters can be determined, subject to all applicable boundary conditions.

The formalism that has been summarized above is somewhat more complicated than we will need for this particular investigation, but it may also find use as a reference point for related studies. Our central purpose here will be to study the response of a resonant medium in the case that the electromagnetic pulse envelope varies significantly on a time scale of an optical cycle. Complications of the model which do not elucidate that particular topic will be set aside in the following sections.

III. SPECIFIC MODEL

One important feature of the model described in Sec. II is its inclusion of an arbitrary orientational distribution of the transition dipoles. With this formalism one can calculate the anisotropic gain distribution that results for arbitrary polarizations of the pump and signal fields [23]. On the other hand, pending some particular application for such polarization effects, they are not required for an initial investigation of few-cycle pulse interactions. Thus it will now be assumed that the medium is orientationally homogeneous, or more specifically that all of the matrix elements of the dipole moment operator are parallel to the linearly polarized electric-field vector.

If the dipole matrix elements of all of the atoms are parallel to the field Eqs. (12)–(17) reduce to the scalar set

$$\left(\frac{\partial}{\partial t}+v\,\frac{\partial}{\partial z}\right)\rho_{21}(v,\omega_\alpha,z,t)$$

$$=-\left[i\omega_\alpha+\frac{i}{\hbar}\,(\mu_{22}-\mu_{11})E(z,t)+\gamma\right]\rho_{21}(v,\omega_\alpha,z,t)$$

$$-\frac{i}{\hbar}\,\mu_{21}E(z,t)[\rho_{22}(v,\omega_\alpha,z,t)-\rho_{11}(v,\omega_\alpha,z,t)], \tag{18}$$

$$\left(\frac{\partial}{\partial t}+v\,\frac{\partial}{\partial z}\right)\rho_{22}(v,\omega_\alpha,z,t)$$

$$=\lambda_2(v,\omega_\alpha,z,t)-\gamma_2\rho_{22}(v,\omega_\alpha,z,t)-\frac{i}{\hbar}\,E(z,t)$$

$$\times[\mu_{12}\rho_{21}(v,\omega_\alpha,z,t)-\mu_{21}\rho_{12}(v,\omega_\alpha,z,t)], \tag{19}$$

$$\left(\frac{\partial}{\partial t}+v\,\frac{\partial}{\partial z}\right)\rho_{11}(v,\omega_\alpha,z,t)$$

$$=\lambda_1(v,\omega_\alpha,z,t)-\gamma_1\rho_{11}(v,\omega_\alpha,z,t)+\gamma_{21}\rho_{22}(v,\omega_\alpha,z,t)$$

$$+\frac{i}{\hbar}\,E(z,t)[\mu_{12}\rho_{21}(v,\omega_\alpha,z,t)-\mu_{21}\rho_{12}(v,\omega_\alpha,z,t)], \tag{20}$$

$$\rho_{12}(v,\omega_\alpha,z,t)=\rho_{21}^*(v,\omega_\alpha,z,t), \tag{21}$$

$$\frac{\partial^2E(z,t)}{\partial z^2}-\mu_1\sigma\,\frac{\partial E(z,t)}{\partial t}-\mu_1\varepsilon_1\,\frac{\partial^2E(z,t)}{\partial t^2}=\mu_1\,\frac{\partial^2P(z,t)}{\partial t^2}, \tag{22}$$

$$P(z,t)=\int_0^\infty\int_{-\infty}^\infty n(v,\omega_\alpha,z,t)[\mu_{11}\rho_{11}(v,\omega_\alpha,z,t)$$

$$+\mu_{21}\rho_{12}(v,\omega_\alpha,z,t)+\mu_{12}\rho_{21}(v,\omega_\alpha,z,t)$$

$$+\mu_{22}\rho_{22}(v,\omega_\alpha,z,t)]dv\;d\omega_\alpha. \tag{23}$$

Formally, this reduction has been achieved by requiring that the density distribution $n(v,\omega_\alpha,\theta,\phi,z,t)$ include a δ-function angular factor. This factor has been multiplied by each of the four density-matrix equations. The resulting equations have been integrated over all angles, and new pump and density matrix variables have been introduced reflecting the integrated form of the old variables. Equations (18)–(23) are still a complete set from which the time and space dependences of the electric field and of the atomic or molecular parameters can in principle be determined.

As a next simplification, it will be assumed that the medium is spectrally homogeneous. If all of the atoms have the same intrinsic center frequency ($\omega_\alpha=\omega_0$), and Doppler effects are unimportant ($v=0$), then Eqs. (18)–(21) and (23) reduce to

$$\frac{\partial}{\partial t}\,\rho_{21}(z,t)=-\left[i\omega_0+\frac{i}{\hbar}\,(\mu_{22}-\mu_{11})E(z,t)+\gamma\right]\rho_{21}(z,t)$$

$$-\frac{i}{\hbar}\,\mu_{21}E(z,t)[\rho_{22}(z,t)-\rho_{11}(z,t)], \tag{24}$$

$$\frac{\partial}{\partial t}\,\rho_{22}(z,t)=\lambda_2(z,t)-\gamma_2\rho_{22}(z,t)-\frac{i}{\hbar}\,E(z,t)$$

$$\times[\mu_{12}\rho_{21}(z,t)-\mu_{21}\rho_{12}(z,t)], \tag{25}$$

$$\frac{\partial}{\partial t}\,\rho_{11}(z,t)=\lambda_1(z,t)-\gamma_1\rho_{11}(z,t)+\gamma_{21}\rho_{22}(z,t)+\frac{i}{\hbar}\,E(z,t)$$

$$\times[\mu_{12}\rho_{21}(z,t)-\mu_{21}\rho_{12}(z,t)], \tag{26}$$

$$\rho_{12}(z,t)=\rho_{21}^*(z,t), \tag{27}$$

$$P(z,t)=n(z,t)[\mu_{11}\rho_{11}(z,t)+\mu_{21}\rho_{12}(z,t)+\mu_{12}\rho_{21}(z,t)$$

$$+\mu_{22}\rho_{22}(z,t)]. \tag{28}$$

This reduction has been achieved by requiring that the density distribution $n(v,\omega_\alpha,z,t)$ include a δ-function factor in the intrinsic center frequency and the velocity. This factor has been multiplied by each of the four density-matrix equations. The resulting equations have been integrated over all frequencies and velocities, and new pump and density-matrix variables have been introduced reflecting the integrated form of the old variables.

It would be usual in a calculation of this type to postulate that the wave functions have parity. In this case the dipole matrix elements μ_{22} and μ_{11} would vanish in Eqs. (24) and (28) and their predecessors. In fact if the rotating-wave approximation were valid, one finds that the terms involving μ_{22} and μ_{11} in Eq. (24) would average to zero even without a parity assumption, and the corresponding terms in Eq. (28) could introduce only a slowly varying (nonoptical frequency) polarization component. Then, with suitable restrictions on the pumping and decay processes, special cases of this model would be compatible with standard homogeneously-broadened-medium density-matrix formulations [24,25]. However, one purpose of this study is to explore the response of a laser medium in cases where the electromagnetic fields vary too quickly for the rotating-wave approximation to be applicable. Thus, it is of interest here to see what sorts of effects usually neglected terms like μ_{22} and μ_{11} might imply.

One of the simplest and most relevant applications of this formalism is to a two-level medium which, prior to the arrival of the electromagnetic pulse, is resting peacefully in its ground state. To explore this case, we will turn off the pumping rates ($\lambda_1=\lambda_2=0$) and specialize the relaxation rates according to $\gamma_1=0$, $\gamma_{21}=\gamma_2$. In this case Eqs. (25) and (26) reduce to

$$\frac{\partial}{\partial t}\,\rho_{22}(z,t)=-\gamma_2\rho_{22}(z,t)-\frac{i}{\hbar}\,E(z,t)[\mu_{12}\rho_{21}(z,t)$$

$$-\mu_{21}\rho_{12}(z,t)], \tag{29}$$

$$\frac{\partial}{\partial t}\,\rho_{11}(z,t)=+\gamma_2\rho_{22}(z,t)+\frac{i}{\hbar}\,E(z,t)[\mu_{12}\rho_{21}(z,t)$$

$$-\mu_{21}\rho_{12}(z,t)]. \tag{30}$$

It is now helpful to introduce a new parameter which combines the off-diagonal density matrix elements with the off-diagonal dipole moment matrix elements $\eta=\mu_{12}\rho_{21}$ [26]. With this substitution Eqs. (24) and (28)–(30) become

$$\frac{\partial}{\partial t}\,\eta(z,t)=-\left[i\omega_0+\frac{i}{\hbar}\,(\mu_{22}-\mu_{11})E(z,t)+\gamma\right]\eta(z,t)$$

$$-\frac{i}{\hbar}\,\mu_{12}\mu_{21}E(z,t)[\rho_{22}(z,t)-\rho_{11}(z,t)], \tag{31}$$

$$\frac{\partial}{\partial t}\rho_{22}(z,t) = -\gamma_2\rho_{22}(z,t) - \frac{i}{\hbar}E(z,t)[\eta(z,t) - \eta^*(z,t)], \tag{32}$$

$$\frac{\partial}{\partial t}\rho_{11}(z,t) = +\gamma_2\rho_{22}(z,t) + \frac{i}{\hbar}E(z,t)[\eta(z,t) - \eta^*(z,t)], \tag{33}$$

$$P(z,t) = n(z,t)[\mu_{11}\rho_{11}(z,t) + \mu_{22}\rho_{22}(z,t) + \eta(z,t) + \eta^*(z,t)], \tag{34}$$

where use has been made of the Hermitian character of the density and dipole moment matrices. An immediate implication of Eqs. (31)–(34) is that the polarization is independent of the phase angle of the dipole moment matrix element μ_{12}, since this element only appears in a product with its complex conjugate. This must at least be true after any effects of initial conditions on the wave functions have died away, and as noted above the medium is assumed to have been unperturbed before the arrival of the electromagnetic pulse.

It is also convenient to introduce the magnitude of the off-diagonal dipole moment matrix element $\mu = |\mu_{12}|$, a normalized off-diagonal density matrix element $p = 2\eta/\mu$, a normalized electric field $A = 2\mu E/\gamma\hbar$, a probability difference $d = \rho_{22} - \rho_{11}$, a probability sum $m = \rho_{22} + \rho_{11}$, and a normalized time $\tau = \gamma t$. With these definitions Eqs. (31)–(34) become

$$\frac{\partial}{\partial\tau}p(z,t) = -\left[1 + i\frac{\omega_0}{\gamma} + i\frac{\mu_{22} - \mu_{11}}{2\mu}A(z,\tau)\right]p(z,\tau) - id(z,\tau)A(z,\tau). \tag{35}$$

$$\frac{\partial}{\partial\tau}d(z,\tau) = -\frac{\gamma_2}{\gamma}[d(z,\tau) + m(z,\tau)] + A(z,\tau)p_i(z,\tau), \tag{36}$$

$$P(z,\tau) = n(z,\tau)\mu\left\{\frac{\mu_{11}}{2\mu}[m(z,\tau) - d(z,\tau)] + \frac{\mu_{22}}{2\mu}[m(z,\tau) + d(z,\tau)] + p_r(z,\tau)\right\}, \tag{37}$$

where the subscripts r and i refer, respectively, to the real and imaginary parts. It may be noted that for a true two-level system in this notation the sum m is always equal to unity. These equations can now be simplified a little further and replaced by the real set

$$\frac{\partial}{\partial\tau}p_i(z,\tau) = -p_i(z,\tau) - [\omega_0' + \mu_d A(z,\tau)]p_r(z,\tau) - A(z,\tau)d(z,\tau), \tag{38}$$

$$\frac{\partial}{\partial\tau}p_r(z,\tau) = -p_r(z,\tau) + [\omega_0' + \mu_d A(z,\tau)]p_i(z,\tau), \tag{39}$$

$$\frac{\partial}{\partial\tau}d(z,\tau) = -\rho[1 + d(z,\tau)] + A(z,\tau)p_i(z,\tau), \tag{40}$$

$$P'(z,\tau) = \mu_{sp} + \mu_d[1 + d(z,\tau)] + p_r(z,\tau), \tag{41}$$

where we have introduced the normalized line-center frequency $\omega_0' = \omega_0/\gamma$ [12], the normalized dipole moment difference $\mu_d = (\mu_{22} - \mu_{11})/(2\mu)$, the normalized lower-state dipole moment $\mu_{\rm sp} = \mu_{11}/\mu$, the normalized decay rate ratio $\rho = \gamma_a/\gamma$, and the normalized polarization $P' = P/(n\mu)$. The symbols p_r and p_i represent the real and imaginary parts of the normalized off-diagonal density matrix element.

In Eq. (41) the static lower state dipole moment is represented by the symbol $\mu_{\rm sp}$. The subscript sp here is intended to stand for spontaneous polarization, and thus $\mu_{\rm sp}$ is the normalized static polarization that remains even when an atom or molecule is in its ground state. In our two-level system this ground-state occupation is represented by the conditions $d(z,\tau) = -1$ and $p_r(z,\tau) = 0$. When $\mu_{\rm sp}$ is nonzero, the inevitable temperature dependence of this residual spontaneous polarization is called pyroelectricity [27], and in cases where the polarization exhibits hysteresis it has been termed ferroelectricity in analogy with the corresponding behavior of ferromagnetic media [28]. For purposes of this discussion a static dipole moment is not essential, and thus the term $\mu_{\rm sp}$ will be dropped in our further discussions.

IV. PARITY APPROXIMATION

A significant mathematical complication of the models developed here is that the dependent variables are functions of both space and time. As the various frequency components that result from any nonlinear interactions will in general have different phase velocities, the rigorous solutions to this model would seem to require the detailed specification of boundary conditions followed by complicated numerical solutions of the governing partial differential equations. The results of such calculations might be too specific to yield general insights into the underlying physics. Fortunately, however, there may be some justification for focusing initially on a much simpler problem.

As discussed above, the shortest pulses in an absolute sense have been obtained at wavelengths around 800 nm in the near-infrared region of the spectrum, and those pulses are several cycles in length. The more interesting cases of pulses that are less than a cycle in length have all occurred in the THz or mm region of the spectrum, where the pulses have been obtained by down shifting from the visible or near infrared. The coherence length, which governs the distance over which harmonics might propagate with the same effective phase velocity as the fundamental frequency components, scales as the wavelength and becomes quite large for far-infrared or submillimeter wavelength experiments. This means that phase matching usually does not present a serious problem in such studies, and nonlinear interactions are likely to be limited by absorption rather than by phase mismatch [29]. As in previous studies, we will focus our interest on a thin slab of material and disregard possible effects of z variations [19,20].

The response of a two-level medium to an arbitrarily varying electromagnetic field is governed by Eqs. (38)–(41). For a localized medium, these equations become

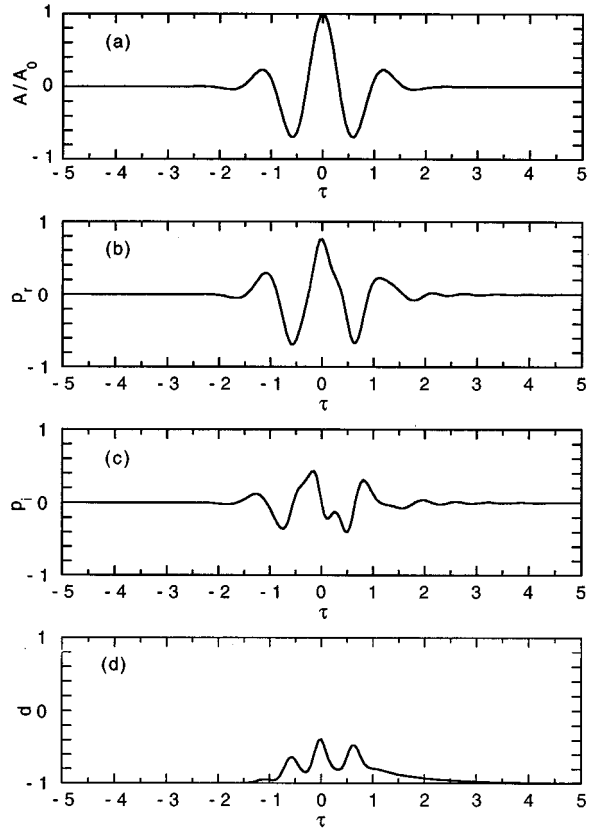

FIG. 1. Solutions of the density-matrix equations that result when a cosinusoidal-Gaussian pulse of width $\tau_0'=1.0$, amplitude $A_0=10.0$, and frequency $\omega'=5.0$ is incident on a system of atoms or molecules characterized by the population decay rate $\rho=1.0$, transition frequency $\omega_0'=10.0$, and dipole moment difference $\mu_d=0.0$. The solutions include (a) the amplitude A/A_0, (b) the real part of the polarization p_r, (c) the imaginary part of the polarization p_i, and (d) the population difference d.

$$\frac{d}{d\tau}p_i(\tau)=-p_i(\tau)-[\omega_0'+\mu_d A(\tau)]p_r(\tau)-A(\tau)d(\tau), \tag{42}$$

$$\frac{d}{d\tau}p_r(\tau)=-p_r(\tau)+[\omega_0'+\mu_d A(\tau)]p_i(\tau), \tag{43}$$

$$\frac{d}{d\tau}d(\tau)=-\rho[1+d(\tau)]+A(\tau)p_i(\tau), \tag{44}$$

$$P'(\tau)=\mu_d[1+d(\tau)]+p_r(\tau), \tag{45}$$

where the static lower state dipole moment μ_{sp} has been dropped.

To explore the implications of Eqs. (42)–(45), it is now necessary to specify the mathematical form of the incident few-cycle electromagnetic pulse. We begin by considering symmetric sinusoidal-Gaussian pulses of the form

$$A(\tau)=A_0\exp(-\tau^2/\tau_0^2)\cos(\omega'\tau), \tag{46}$$

where A_0 is the normalized pulse-envelope amplitude, τ_0 is the normalized $1/e$ half-width in time of the electric-field pulse envelope, and ω' is the normalized frequency of the

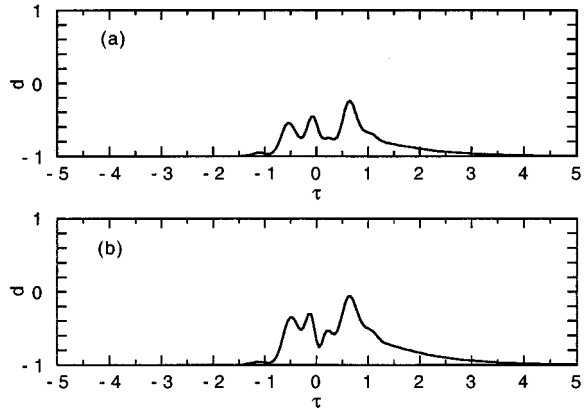

FIG. 2. Population difference solutions for nonzero values of the dipole moment difference μ_d including (a) $\mu_d=0.2$ and (b) $\mu_d=0.5$. In these cases the population difference has higher harmonics of the underlying field frequency.

underlying field oscillations. This Gaussian form for the pulse envelope function is not chosen here for any compelling physical reason, and other pulse shapes might be more appropriate for specific practical applications. The Gaussian envelope does, however, resemble the pulses seen in some systems, and this is one of several shapes that have been employed in pulse propagation studies. The mathematical form of Eq. (46) does not, of course, mean that the pulses actually look like modulated sine waves. For $\omega'<1/\tau_0$, little oscillation occurs during the Gaussian pulse envelope, and the pulse would look more like a simple half-cycle Gaussian.

With suitable parameter choices, Eqs. (42)–(46) may be solved for the interaction of a range of very short pulses with two-level media. As a first step, we will consider briefly some of the consequences of not making the parity approximation. A typical input cosinusoidal-Gaussian laser pulse $A(\tau)$ is shown in Fig. 1(a). In this case the pulse parameters include the normalized amplitude $A_0=10.0$, the normalized width $\tau_0=1.0$, and the normalized frequency $\omega'=5.0$. Figure 1(b) shows the real part of the normalized polarization p_r, that results when the pulse of Fig. 1(a) is incident on a system of atoms characterized by the normalized population decay rate $\rho=1.0$ and the normalized intrinsic transition frequency $\omega_0'=10.0$. The corresponding values of the imaginary part of the polarization p_i and the population difference $d(\tau)$ are shown in Figs. 1(c) and 1(d), respectively. In this example the normalized dipole moment difference μ_d is set equal to zero, which is the appropriate value if the wave functions have parity (or if $\mu_{22}=\mu_{11}$). We see that for these values the population of the upper state is increased sharply whenever the field has either a positive or a negative maximum, and this fact will be discussed further below. The slight general asymmetry in Fig. 1(d) indicates some longer-term upper-state population accumulation.

If the wave functions lack parity, the response of the medium becomes more complex. Figure 2(a) shows the time-dependent population difference for the same parameter values as Fig. 1(d), except that in Fig. 2(a) the dipole moment difference is $\mu_d=0.2$. In this case the population difference has more structure, including the gradual development of a higher harmonic of the underlying electromagnetic field fre-

quency. This behavior should probably not be considered unexpected, since in both Eqs. (42) and (43) the factor μ_d is seen to introduce additional nonlinearities to the model. If the dipole moment difference is increased further, the response becomes still more complicated, and an example is shown in Fig. 2(b) for the value $\mu_d = 0.5$. It is clear that one area of potential interest relating to the parameter μ_d would be nonlinear optics with few-cycle pulses.

As noted above, most studies of two-level atoms assume that the wave functions do have parity. Thus, for simplicity we will now set μ_d equal to zero for the remainder of this study. In this limit Eqs. (42)–(45) reduce to the simpler model

$$\frac{d}{d\tau} p_i(\tau) = -p_i(\tau) - \omega_0' p_r(\tau) - A(\tau) d(\tau), \tag{47}$$

$$\frac{d}{d\tau} p_r(\tau) = -p_r(\tau) + \omega_0' p_i(\tau), \tag{48}$$

$$\frac{d}{d\tau} d(\tau) = -\rho[1 + d\tau] + A(\tau) p_i(\tau), \tag{49}$$

$$P'(\tau) = p_r(\tau). \tag{50}$$

This model will be the basis for all of the following considerations.

V. RATE-EQUATION-LIKE APPROXIMATION

An interesting feature of the results noted in the previous section is that if μ_d is equal to zero ($\mu_{22} = \mu_{11}$), the upper-state population tends to increase whenever the field has either a positive or a negative maximum. This curious behavior is not a consequence of the particular cosine-Gaussian pulse shape that was adopted for Fig. 1(a). To show this, the corresponding results with a sine-Gaussian pulse are presented in Fig. 3. The sine-Gaussian input pulse is shown in Fig. 3(a) and the associated population difference is shown in Fig. 3(d) using all of same parameter values as in Fig. 1. It is clear from this comparison that, independent of the relative phases of the wave and its envelope, the population is enhanced by both the positive and negative polarity phases of the input pulse.

The polarity independence can have a fairly simple interpretation for certain operating conditions. For example, we may suppose that the incident field pulse has a carrier wave that, like Eq. (46), is cosinusoidal in form. If the field is intense (as in Fig. 1, where $A_0 = 10.0$), and the transition frequency is large (as in Fig. 1, where $\omega_0' = 10.0$), it is helpful to first guess that the real polarization will also be somewhat cosinusoidal [which Fig. 1(b) shows is the case]. It then follows from Eq. (47) that the imaginary polarization must be somewhat sinusoidal [which Fig. 1(c) shows it is]. Then it is clear from Eq. (48) that, as postulated, the real polarization is indeed somewhat cosinusoidal. The driving term in Eq. (49) must now be in the form of a cosine times a sine, which corresponds to a sine at twice the original frequency. Finally, Eq. (49) implies that the population difference will be modulated according to a cosine function at twice the input frequency, and this is exactly what is seen in Fig. 1(d). Similar

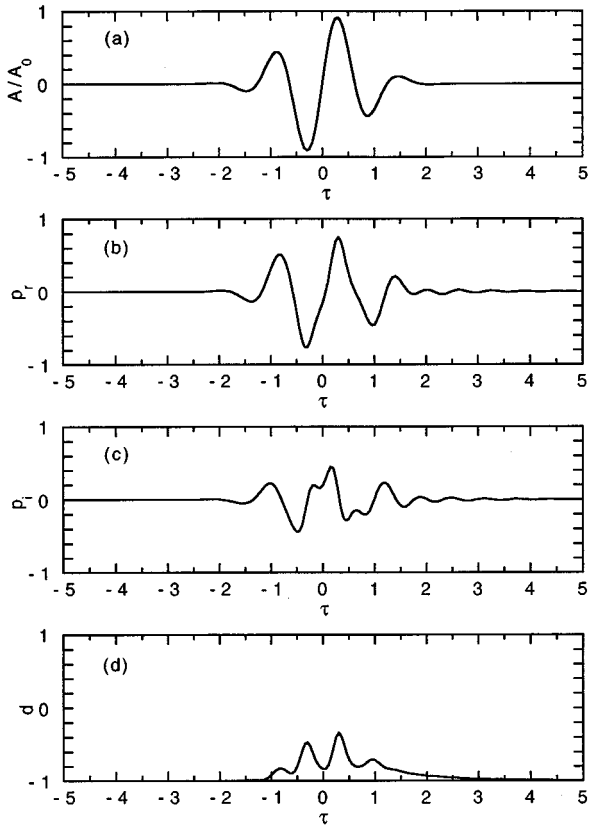

FIG. 3. Solutions for a sinusoidal-Gaussian input pulse with the same parameter values as Fig. 1. As in Fig. 1 the population difference is enhanced by both the positive and negative polarity phases of the input pulse.

arguments provide an explanation for the double-frequency oscillations of the population difference in Fig. 3(d).

The above interpretation relied on the fact that both A_0 and ω_0' are large in this example. Interestingly, very similar results are also obtained under what would seem to be quite different conditions. We first observe that the real and imaginary polarization components tend to relax to zero in a time on the order of unity in these normalized units. If the driving terms in these equations vary slowly enough on this time scale, it becomes a good approximation to set the time derivatives in the polarization equations equal to zero. For conventional longer-wavelength pulses, for example, this condition might sometimes be well satisfied. This type of simplification is often known as adiabatic elimination, and in conventional rotating-wave-approximated optical systems the elimination of polarization variables is more commonly called the rate-equation approximation. A systematic study of this type of approximation, and its more accurate versions, has recently been reported [30].

The applicability of the rate-equation-like approximation that has just been described may be shown by means of an example. In Fig. 4 is a plot of the equation solutions for the same conditions as for Fig. 1, except that the input pulse for Fig. 4 is much longer and weaker. In particular, the pulse amplitude is here reduced to $A_0 = 1.0$, the pulse width is $\tau_0 = 10.0$, and the frequency is $\omega' = 0.5$. It may be seen that in this case the real and imaginary polarization components are

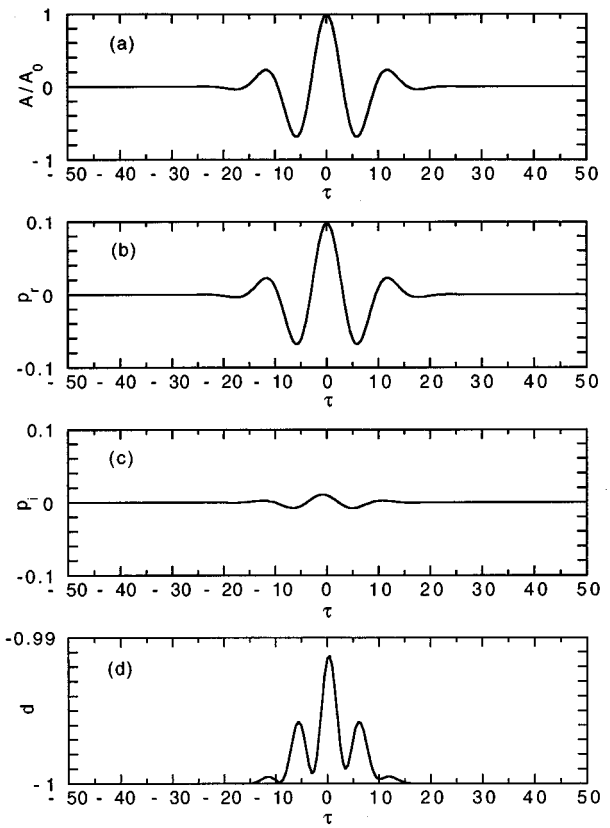

FIG. 4. Solutions for a cosinusoidal-Gaussian input pulse with the same parameter values as Fig. 1, except that the pulse amplitude is here reduced from $A_0=10.0$ to $A_0=1.0$, the width is increased from $\tau_0=1.0$ to $\tau_0=10.0$, and the frequency is reduced from $\omega'=5$ to $\omega'=0.5$. In contrast to Fig. 1, the polarization components are both in phase with the field.

both approximately in phase with the driving field, and this is characteristic of adiabatic following behavior. Interestingly, the population difference is still modulated at twice the input frequency, even though the conditions here are very different.

If the left-hand sides of Eqs. (47) and (48) are set equal to zero, the resulting algebraic equations can be solved for the polarization components. The results are

$$p_i(\tau)=-\frac{A(\tau)d(\tau)}{1+\omega_0'^2}, \tag{51}$$

$$p_r(\tau)=-\frac{\omega_0' A(\tau)d(\tau)}{1+\omega_0'^2}. \tag{52}$$

When Eq. (51) is substituted into Eq. (49), one obtains the differential equation

$$\frac{d}{d\tau}d(\tau)+\left(\frac{A^2(\tau)}{1+\omega_0'^2}+\rho\right)d(\tau)=-\rho. \tag{53}$$

The formal integral of this equation can be written as [31]

$$d(\tau)=-\rho\exp\left[-\int^{\tau}\left(\frac{A^2(\tau')}{1+\omega_0'^2}+\rho\right)d\tau'\right]\times\int^{\tau}\exp\left[\int^{\tau'}\left(\frac{A^2(\tau'')}{1+\omega_0'^2}+\rho\right)d\tau''\right]d\tau'+C\exp\left[-\int^{\tau}\left(\frac{A^2(\tau')}{1+\omega_0'^2}+\rho\right)d\tau'\right], \tag{54}$$

where C is an integration constant.

For some forms for the input pulse it would be possible to simplify Eq. (54) analytically. However, the important aspect of this equation for our present purposes is that the population difference depends only on the square of the electric-field amplitude rather than on the amplitude itself. In particular, the population difference is still driven upward during both phases of the input field, in agreement with the plots shown in Fig. 4(d). This result is analogous to the more conventional rate-equation approximations in which the populations are driven by the electromagnetic intensities rather than the fields. The distinction between these short-pulse results and the conventional rate-equation approximation is, however, very significant. The intensity is not proportional to the field squared, but only to the time average of that quantity. This distinction has arisen in the present discussion because we have not yet considered the rotating-wave approximation. In contrast to all previous treatments we are making (or at least considering making) the rate-equation-like approximation before the rotating-wave approximation. Thus we wish to emphasize that these two approximations rest on different assumptions, and, at least in principle, they are independent of each other. The rate-equation (-like) approximation requires that the field amplitude envelope (or just the instantaneous field) and the populations vary slowly compared to the coherence decay time. On the other hand, the rotating-wave approximation, as will be discussed below, requires that the fields not be too strong and that the polarization and population components vary slowly compared to an optical cycle. With very short pulses the coherence decay time may be greater than or less than an optical cycle, and thus these two approximations may become valid or invalid independently of each other.

VI. ROTATING-WAVE APPROXIMATION

One of the most basic approximations in dealing with the interaction of light with atoms is, for historical reasons, known as the rotating-wave approximation, and this approximation has long been recognized [32]. The rotating-wave approximation is generally found to be valid as long as the optical fields are not too intense, and the polarization and population components do not vary significantly within an optical cycle. Thus, especially in cases of very high intensities [13] or very short pulses [19,20], the validity of the approximation may be in doubt. Our emphasis here is on electromagnetic pulses that are at most a few cycles in length, and it is appropriate to consider the adaptations of our model that might be necessary to test this approximation.

As a starting point, we rewrite Eqs. (47)–(49) in the form

$$\frac{d}{d\tau}p(\tau)=-p(\tau)-i\omega_0' p(\tau)-iA(\tau)d(\tau), \tag{55}$$

$$\frac{d}{d\tau}d(\tau)=-\rho[1+d(\tau)]+A(\tau)p_i(\tau), \tag{56}$$

As noted above, we could begin here instead with Eqs. (42)–(44), in which the parity approximation has not been made, but the rotating-wave approximation as generalized to our non-parity-approximated model is readily seen to eliminate the effects of the nonzero diagonal dipole matrix elements. Thus, the inclusion of these dipole terms would obscure other fundamental questions about the effects of the approximation, and for brevity we start with the parity-approximated model.

To be specific, we assume that the field and polarization can be written in the forms

$$A(\tau)=A_0(\tau)\cos(\omega'\tau)$$
$$=\frac{A_0(\tau)}{2}[\exp(i\omega'\tau)+\exp(-i\omega'\tau)], \tag{57}$$

$$p(\tau)=p_0(\tau)\exp(-i\omega'\tau). \tag{58}$$

With these substitutions, and division by the negative exponential, Eqs. (55) and (56) take the forms

$$\frac{d}{d\tau}p_0(\tau)=-p_0(\tau)+i(\omega'-\omega_0')p_0(\tau)$$
$$-i\frac{A_0(\tau)d(\tau)}{2}[\exp(2i\omega'\tau)+1], \tag{59}$$

$$\frac{d}{d\tau}d(\tau)=-\rho[1+d(\tau)]+\frac{A_0(\tau)p_{0i}(\tau)}{4}[2+\exp(2i\omega'\tau)$$
$$+\exp(-2i\omega'\tau)]+i\frac{A_0(\tau)p_{0r}(\tau)}{4}[\exp(2i\omega'\tau)$$
$$-\exp(-2i\omega'\tau)], \tag{60}$$

where the subscripts r and i on the polarization amplitude again denote the real and imaginary parts.

The rotating-wave approximation now consists of time averaging and thus dropping all the oscillating exponential terms. Within this approximation Eqs. (59) and (60) can be written in the real forms

$$\frac{d}{d\tau}p_{0i}(\tau)=-p_{0i}(\tau)+(\omega'-\omega_0')p_{0r}(\tau)-\frac{A_0(\tau)d(\tau)}{2}, \tag{61}$$

$$\frac{d}{d\tau}p_{0r}(\tau)=-p_{0r}(\tau)-(\omega'-\omega_0')p_{0i}(\tau), \tag{62}$$

$$\frac{d}{d\tau}d(\tau)=-\rho[1+d(\tau)]+\frac{A_0(\tau)p_{0i}(\tau)}{2}. \tag{63}$$

The advantage of the rotating-wave approximation for long-pulse or cw electromagnetic waves is, of course, that the rapidly varying functions have all been eliminated from the equations, and in seeking either analytical or numerical solutions one need only be concerned in these equations with the envelope functions for the electric field and polarization. For the very short pulse envelopes of interest here, the validity of the rotating-wave approximation is not always assured.

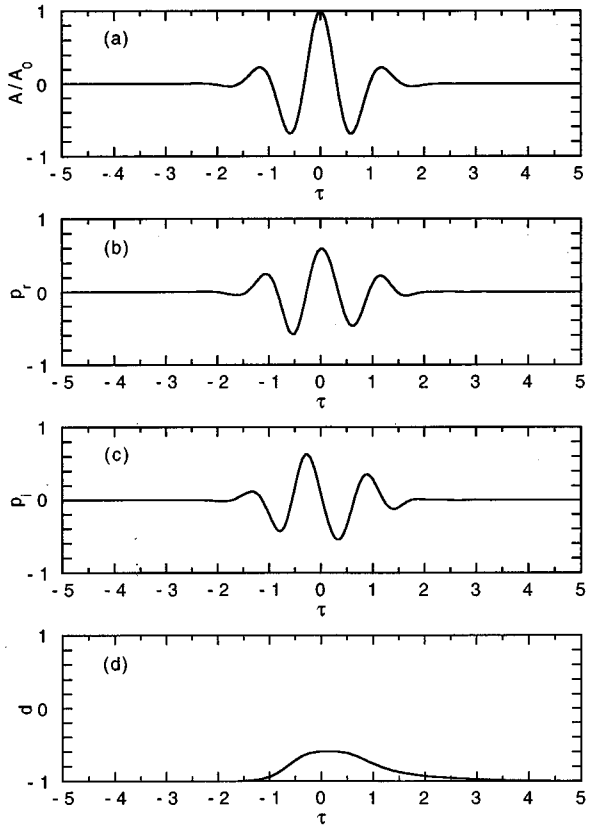

FIG. 5. Solutions for the same parameter values as Fig. 1 but with the rotating-wave-approximation. This approximation eliminates frequency harmonics from the variables.

It is interesting to note the very close resemblance in form between Eqs. (47)–(49) (before the approximation) and Eqs. (61)–(63) (after the approximation). The most fundamental difference is that in the general set one is dealing with the absolute frequency of the transition ω_0', whereas in the approximate set only the difference between that frequency and the assumed carrier frequency $(\omega'-\omega_0')$ appears. Because of this similarity the solution methods for the two sets are identical. To obtain a rigorous comparison between the predictions of the two models, it is only necessary in the approximate set to transform the results back to the original field variables.

A set of solutions to Eqs. (61)–(63) are given in Fig. 5. In this figure the plotted variables again include (a) the input field amplitude $A(\tau)$, (b) the real part of the polarization $p_r(\tau)$, (c) the imaginary part of the polarization $p_i(\tau)$, and (d) the population difference $d(\tau)$. The parameters used in these plots include the peak amplitude $A_0=10.0$, the width $\tau_0=1.0$, the population decay rate $\rho=1.0$, the frequency $\omega'=5.0$, and the transition frequency $\omega_0'=10.0$. Since these parameter values are the same as those employed in obtaining Fig. 1, the two sets of results may be compared directly.

It is clear from this comparison that the rotating-wave approximation eliminates frequency harmonics from the variables, and its simplifying effects on the population difference are particularly conspicuous. Interestingly, the rate-equation-like approximation discussed above can retain this faster structure. However, for many values of the governing parameters in few-cycle interactions neither of these approximations permits an accurate representation of the actual population and polarization dynamics.

VII. OTHER PARAMETER VARIATIONS

For any potentially realistic model for the interaction of light with atoms, there must be many parameters to match with experimental conditions. That is true in the present case, but except for the diagonal dipole matrix elements we have not focused on the consequences of different values for these parameters. In this section we will consider some of these parameters very briefly, and for the most part it will be straightforward to interpret their implications physically.

One parameter that can be understood almost independently from other aspects of the model is the decay rate ratio $\rho = \gamma_2 / \gamma$. To illustrate the effects of ρ, we compare typical solutions of Eqs. (47)–(49) that differ only in the adopted value for that parameter. The normalized amplitude pulse and other parameters used in Fig. 1 are also the basis for the results given in Fig. 6. The decay rate ratio $\rho = 1$ was used in obtaining the population difference curve in Fig. 1(d), and the corresponding population difference results for smaller values of ρ including $\rho = 0.5$, 0.2, 0.1, and 0.0 are plotted in Fig. 6. These results have the straightforward interpretation that with smaller values of the decay rate ratio the population collects in the upper state for a longer period of time. In many practical media the population decay lifetime is much longer than the phase-coherence time, so this simplest possible requirement $\rho = 0$ would often be very realistic for short-pulse applications. If $\rho = 0$, Eq. (49) may be replaced by

$$\frac{d}{d\tau} d(\tau) = A(\tau) p_i(\tau). \tag{64}$$

Another parameter of interest is the transition frequency. To illustrate the effects of ω_0', we compare typical solutions of Eqs. (47)–(49) that differ mainly in the adopted value for that parameter. The normalized amplitude pulse and other parameters used in Fig. 1 are retained. The transition frequency $\omega_0' = 10.0$ was used in obtaining the results shown in Fig. 1, and the corresponding population difference results for the smaller value $\omega_0' = 5.0$ are plotted in Fig. 7. In this case the driving field is essentially at the resonance for the transition, and to obtain a comparable vertical scale the field amplitude has been reduced from 10.0 to 2.0. We see that at resonance there is a longer delay in the development and decay of the polarization oscillations, and also a much greater sensitivity to the input field.

Another parameter of particular interest is the pulse amplitude. This is one of the parameters that bears on the validity of using a two-level model for a real optical medium. To illustrate the effects of A_0, we compare typical solutions of Eqs. (47)–(49) that differ in the adopted value for that parameter. The other parameters used in Fig. 1 are retained. The amplitude $A_0 = 10.0$ was used in obtaining the results shown in Fig. 1, and the corresponding population difference results for the smaller values $A_0 = 5.0$ and 2.0 are plotted in Fig. 8. It is clear from the figure that, as one would expect, smaller values of the amplitude leave the upper state with a lower population.

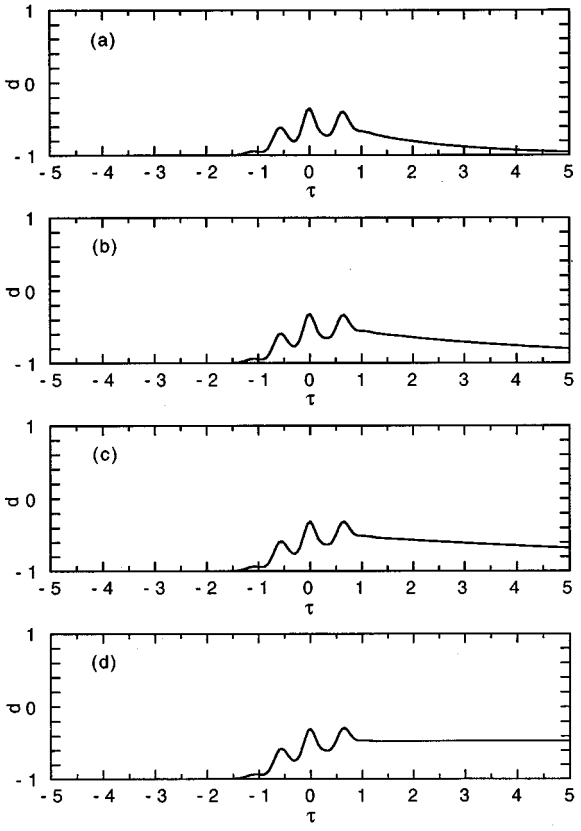

FIG. 6. Solutions for the population difference using the same parameter values as Fig. 1, except that the decay rate ratio takes on the values (a) $\rho = 0.5$, (b) $\rho = 0.2$, (c) $\rho = 0.1$, and $\rho = 0.0$. With smaller values of the decay rate ratio, the population collects in the upper state.

VIII. DISCUSSION

In this work we have undertaken a systematic investigation of the interaction of very short electromagnetic pulses with two-level media. The pulses under consideration are only a few cycles in length, or may even be less than a single cycle. For pulses of such lengths many of the standard techniques and approximations are of doubtful validity. As a foundation for this work and possible future studies, a formal semiclassical model was briefly developed including most of the line broadening and decay processes that one would ever likely be interested in for amplifier or absorber investigations.

For the detailed results discussed here, we have focused on the special case of a two-level absorber in which the lower level of the transition is the ground state. With this example together with the more general models, it would be straightforward to compute and sometimes intuit the behav-

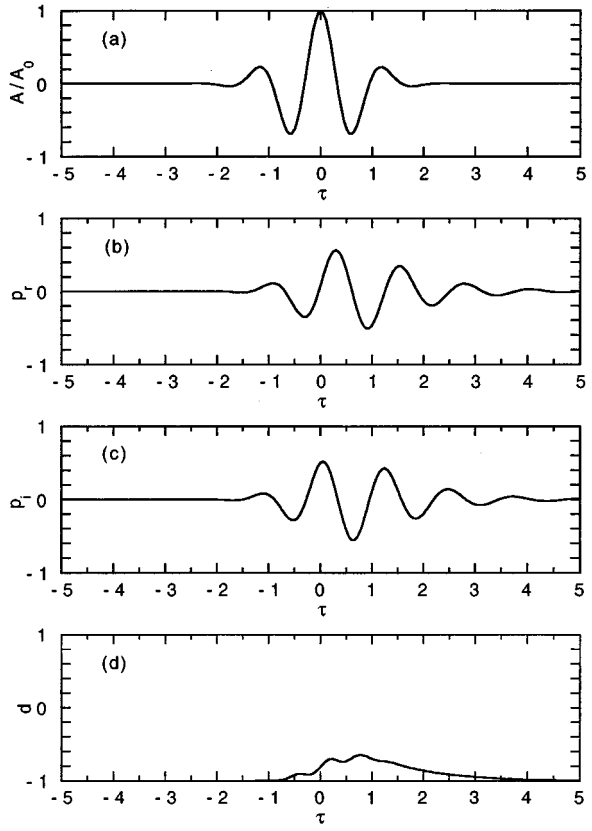

FIG. 7. Solutions for the population difference using the same parameter values as Fig. 1, except that the transition frequency has been reduced from $\omega_0'=10.0$ to $\omega_0'=5.0$ (near a resonance with the pulse), and the pulse amplitude has been reduced from $A_0=10.0$ to $A_0=2.0$. The development and decay of the variables is slower near resonance, and sensitivity to the input field is increased.

ior of other cases of potential interest. Among the results presented here are a semiclassical formalism for interactions with media in which the diagonal elements of the dipole moment matrix are not equal to zero. For very short pulses these elements may have a strong effect on the polarization and population response of a two-level medium, while for longer pulses describable with the rotating-wave approximation these matrix elements have no effect on the interaction dynamics.

One of the most common approximations in studying the interaction of electromagnetic fields with atoms is commonly referred to as the rate-equation approximation. In this approximation, polarization variables are adiabatically eliminated from the overall governing model. Always in the past this approximation has been made subsequent to the even more universally employed rotating-wave approximation. However, in the context of interactions with very short

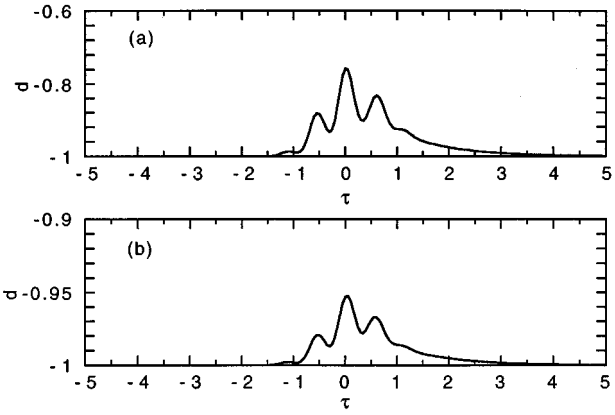

FIG. 8. Solutions for the population difference using the same parameter values as Fig. 1, except that the input pulse amplitude is reduced to (a) $A_0=5.0$ and (b) $A_0=2.0$. As expected, smaller pulses lead to lower populations with little other consequence.

pulses, both of these approximations may fail to give a satisfactory description of the interactions. We have suggested that a rate-equation-like approximation may sometimes be applicable even when the rotating-wave approximation is not. The standard rate-equation approximation requires that the field envelope and populations vary negligibly within the phase-coherence time, while the rotating-wave approximation requires that the polarization amplitudes and populations vary negligibly within an optical cycle. In very short-pulse interactions the coherence time might be greater than a period of the electromagnetic field as in typical optical cases, or it might in principle be less than the period for THz or other very low-frequency waves. Our rate-equation-like approximation involves the elimination of the polarization variables in a model which retains the absolute amplitude and phase of the optical pulse.

We have also reported solutions for the problem of the interaction of electromagnetic pulses with atoms for cases in which the rotating-wave approximation is and is not applied. Without this approximation the population difference typically has substantial variations at twice the frequency of the driving field, and such extra harmonic content also appears in the polarization components. With the application of the rotating-wave approximation the fine structure necessarily vanishes, and this discrepancy confirms the invalidity of the rotating-wave approximation for seemingly reasonable parameter values in very short-pulse systems.

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