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Adiabatic-Following Approximation

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The nonlinear response of an atom to a near-resonant light pulse is studied using a novel approximation scheme. In first order, the approximate solution reduces to the well-known rate equations. The second-order approximation contains Grischkowsky's adiabatic-following approximation. In each order, the approximate solution of the Bloch equations is presented with a closed-form expression for the error that can be used to investigate its range of validity.

INTRODUCTION

The *resonant* interaction of short coherent light pulses with matter has recently generated much experimental[1] and theoretical interest.[2] This article presents a theoretical analysis of some nonlinear effects which can occur when a *near-resonant* coherent light pulse interacts with matter. The work that follows was motivated by a series of elegant experiments involving near-resonant pulse propagation which were performed by Grischkowsky.[3-5] The experiments of Refs. 3–5 were adequately explained by an intuitive vector model. It follows from the vector model that when the effective field changes direction slowly compared with the precessional period of the Bloch vector and when the pulse is sufficiently short that relaxation times T_1 and T_2' can be neglected, the Bloch vector remains parallel to the effective field. If the two conditions stated above are satisfied, then the adiabatic following approximation is good and the atomic dipole moment induced by the off-resonant light pulse is a relatively simple nonlinear function of the light pulse's amplitude. Adiabatic following can be thought of as an optical analog of spin locking.[6,7] The goal of this article is to present an analytic derivation of the adiabatic following approximation from the Bloch equations which describe the interaction of a two-level atom with any coherent light pulse. The derivation of the adiabatic-following equations will also provide a closed-form expression for the error incurred in this approximation.

In order to describe the response of an atom to an off-resonant light pulse, a new approximation scheme is introduced. If pursued indefinitely, the *Ansatz* presented below would provide a complete description of the response of an atom to an off-resonant pulse. As usually occurs in such expansions, only the first few terms are of simple enough form to be useful.

FORMULATION

Consider an atom that is perturbed by a light pulse which propagates in the z direction and has an electric field of the form

$$\vec{E}(z,t)=\mathcal{E}(z,t)\{\hat{e}_x\cos[\omega(t-z/c)-\phi(z,t)] + \hat{e}_y\sin[\omega(t-z/c)-\phi(z,t)]\}. \quad (1)$$

If the pulse frequency ω is in near-resonance with an isolated pair of levels and other levels are not populated, any state of the atom can be described by the wavefunction

$$\Psi=a(t)\psi_a(\vec{x})+b(t)\psi_b(\vec{x}), \quad (2)$$

where ψ_a and ψ_b are eigenfunctions of the unperturbed atomic Hamiltonian. Instead of solving the Schrödinger equation for the time-dependent coefficients $a(t)$ and $b(t)$ it is useful to introduce three real variables[8] X, Y, and Z, which are related to the quantum amplitudes according to

$$(X-iY)\exp -i[\omega(t-z/c)-\phi]\equiv 2ab^*, \quad (3a)$$

$$Z\equiv aa^*-bb^*. \quad (3b)$$

In terms of these variables the expectation of the atom's dipole moment operator and energy are

$$\langle\vec{\mu}_{\rm op}\rangle=\mu\,\mathrm{Re}\{(\hat{e}_x+i\hat{e}_y)(X-iY)\exp[-i\omega(t-z/c)+i\phi]\} \quad (4a)$$

and

$$\langle\mathcal{H}_{\rm atom}\rangle=(\hbar\Omega/2)Z, \quad (4b)$$

where $\Omega=(E_a-E_b)/\hbar$ is the transition frequency and μ the dipole moment matrix element between the levels a and b.

From Eq. (4a) it is seen that the polarization which would result from a collection of N atoms per cubic centimeter is given by

$$\vec{P}(z,t)=N\mu\,\mathrm{Re}\left\{(\hat{e}_x+i\hat{e}_y)\int_{-\infty}^{\infty}(X-iY)g(\Omega-\omega)\,d\Omega \times\exp[-i\omega(t-z/c)+i\phi]\right\}, \tag{5}$$

where the integral over the inhomogeneous line-shape function $g(\Omega-\omega)$ takes into account the possibility of a distribution of transition frequencies Ω. In the case of a gas the inhomogeneous broadening would result from the Doppler effect.

Feynman, Vernon, and Hellwarth[9] have introduced three real variables r_1, r_2, and r_3 that differ from X, Y, and Z by a simple rotation about the Z axis,

$$(X-iY)\exp -i[\omega(t-z/c)-\phi]=r_1-ir_2\,, \tag{6a}$$

$$Z=r_3\,. \tag{6b}$$

The variables u, v, and W of McCall and Hahn[1] are related to X, Y, and Z as follows:

$$u=\tfrac{1}{2}N\mu X\,, \tag{7a}$$

$$v=-\tfrac{1}{2}N\mu Y\,, \tag{7b}$$

$$W=\tfrac{1}{2}N\hbar\Omega Z\,. \tag{7c}$$

If relaxation processes are absent, conservation of probability requires that

$$aa^*+bb^*=1=X^2+Y^2+Z^2\,. \tag{8}$$

When written in terms of X, Y, and Z the Schrödinger equation has the form

$$\dot{X}-i\dot{Y}=-[1/T_2'+i(\Delta+\dot{\phi})](X-iY)-i(\mu\mathcal{E}/\hbar)Z\,, \tag{9a}$$

$$\dot{Z}=-(\mu\mathcal{E}/\hbar)Y-(Z-Z_{\mathrm{eq}})/T_1\,, \tag{9b}$$

where $\Delta=\Omega-\omega$ is the difference between an atom's transition frequency and the laser pulse's frequency. Equations (9a) and (9b) are referred to as the optical Bloch equations. The homogeneous transverse relaxation time T_2' and longitudinal relaxation time T_1 have been introduced phenomenologically.

Propagation of the light pulse through a medium consisting of a dilute collection of N atoms per cubic centimeter is described by the reduced wave equation[8]

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\mathcal{E}(z,t)e^{i\phi(z,t)}=\frac{2\pi}{c}\left(i\omega\mathcal{P}(z,t)-2\,\frac{\partial\mathcal{P}(z,t)}{\partial t}\right), \tag{10a}$$

where

$$\mathcal{P}(z,t)=N\mu\int_{-\infty}^{\infty}[X(z,t,\Delta)-iY(z,t,\Delta)]g(\Delta)d\Delta e^{i\phi(z,t)}\,. \tag{10b}$$

The derivation of the approximate reduced wave equation from the exact second-order equation is discussed in Refs. 1 and 2. This derivation requires that the amplitude and phase of the light pulse varies little over a distance of a wavelength and in the time of an optical period. In the analysis of the atomic response to an off-resonant light pulse that appears below, the rate of change of the amplitude and phase of the light pulse is compared with the amount off-resonance Δ, not the optical frequency ω. The fact that Δ/ω is small (it is about 10^{-3} in the experiment of Ref. 3) indicates that higher-order derivatives may be kept in the work that follows. The term involving $\partial\mathcal{P}/\partial t$ on the right-hand side of Eq. (10a) is usually neglected when discussing resonant pulse propagation. However, when discussing near-resonant pulse propagation it is not as good an approximation to neglect $\partial\mathcal{P}/\partial t$ when compared with $i\omega\mathcal{P}$. For the near-resonant case, the ratio of the two terms will be of the order of Δ/ω. The general description of pulse propagation requires a simultaneous solution of Eqs. (9a), (9b), (10a), and (10b). An adequate description of most experiments can be obtained with the initial conditions

$$X(z,-\infty,\Delta)-iY(z,-\infty,\Delta)=0\,, \tag{11a}$$

$$Z(z,-\infty,\Delta)=\pm 1\,. \tag{11b}$$

The positive sign in Eq. (11b) refers to an atom prepared in its upper state; the negative sign refers to an atom prepared in its ground state.

VECTOR MODEL

Since the primary goal of this article is to analytically derive the adiabatic-following approximation from the Bloch equations, it seems appropriate to begin by reviewing its derivation from the vector model.[3] The optical Bloch equations of Eqs. (9a) and (9b) can be written in the form

$$\dot{\vec{R}}=\vec{\Omega}\times\vec{R}\,, \tag{12}$$

when the relaxation times T_1 and T_2' are very long compared with the pulse duration. The Bloch vector is given by

$$\vec{R}=X\hat{e}_x+Y\hat{e}_y+Z\hat{e}_z\,, \tag{13}$$

and the effective field is

$$\vec{\Omega}(t)=-[\mu\mathcal{E}(t)/\hbar]\hat{e}_x+\Delta\hat{e}_z\,, \tag{14}$$

when the frequency modulation $\dot{\phi}$ is negligible.

Equation (12) has the geometric interpretation that the Bloch vector tries to precess about the effective field as $\vec{\Omega}$ varies in both magnitude and direction. The instantaneous precession frequency of $\vec{R}$ about $\vec{\Omega}$ is

$$|\vec{\Omega}(t)| = \{[\mu \mathcal{E}(t)/\hbar]^2 + \Delta^2\}^{1/2}. \tag{15}$$

When the amount off-resonance $|\Delta|$ is large enough, the precession frequency of the Bloch vector will be large compared with the rate of change of the effective field. For such large $|\Delta|$ it is a good approximation to assume that the Bloch vector remains parallel or antiparallel to the effective field as it moves adiabatically. Figure 1 illustrates the motion of the Bloch vector when Δ (which is chosen negative in the figure) is so large that this adiabatic-following approximation is good. In this limit the angle α between the Bloch vector and the effective field will remain very small. The fact that the Bloch vector is essentially parallel to the effective field in this limit enables one to immediately write down expressions for X and Z from the figure. Using simple geometry, it follows that

$$X = \left(\frac{\mu \mathcal{E}}{\hbar}\right)\left(\frac{\Delta}{|\Delta|}\right)\frac{1}{[(\mu\mathcal{E}/\hbar)^2 + \Delta^2]^{1/2}}, \tag{16a}$$

$$Z = \frac{-|\Delta|}{[(\mu\mathcal{E}/\hbar)^2 + \Delta^2]^{1/2}}. \tag{16b}$$

The signs in Eqs. (16a) and (16b) would be reversed if it had been assumed that $\vec{R}(z, -\infty, \Delta) = +\hat{e}_z$ instead of $\vec{R}(z, -\infty, \Delta) = -\hat{e}_z$.

Of course the Bloch vector cannot remain exactly parallel to the effective field because this would make the right-hand side of Eq. (12) identically zero and the Bloch vector would not be able to change as the direction of $\vec{\Omega}$ changed. This means that the angle α must be finite, although small, and the Y component of the Bloch vector is not exactly equal to zero. It is a shortcoming of the geometric derivation of the adiabatic-following approximation that it is not possible to derive an expression for Y from it. This failing will be corrected in the analytical derivation which appears below. An expression for the Y component of the Bloch vector can be deduced in a self-consistent manner by substituting Eq. (16a) into Eq. (9a) when $T_2' = \infty$ and $\dot{\phi} = 0$.[5] The resulting expression for Y is

$$Y = -\frac{d}{dt}\left(\frac{\mu\mathcal{E}}{\hbar}\right)\frac{|\Delta|}{[(\mu\mathcal{E}/\hbar)^2 + \Delta^2]^{3/2}}. \tag{16c}$$

This expression along with Eq. (16b) satisfies Eq. (9b) with $T_1 = \infty$.

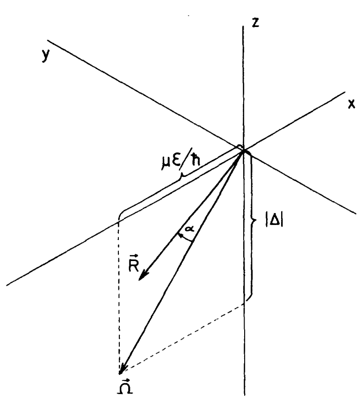

FIG. 1. Precession of the Bloch vector $\vec{R}$ about the effective field $\vec{\Omega}$. When the requirements of adiabatic following are satisfied, the angle α is small and the Bloch vector is essentially parallel to the effective field.

APPROXIMATION SCHEME

Equations (9a), (9b), (11a), and (11b) can be recast as integral equations of the form

$$[X(t) - iY(t)]e^{i\phi(t)} = -i\int_{-\infty}^{t}[\mu\mathcal{E}(t')e^{i\phi(t')}/\hbar]Z(t') \times \exp-(1/T_2' + i\Delta)(t-t')\,dt', \tag{17a}$$

$$Z(t) = Z(-\infty) - \int_{-\infty}^{t}[\mu\mathcal{E}(t')/\hbar]Y(t')\exp-(t-t')/T_1\,dt', \tag{17b}$$

where, for the sake of brevity, the dependence of the variables X, Y, and Z on z and Δ and the dependence of $\mathcal{E}$ and ϕ on z is not explicitly indicated.

Changing the variable of integration to $x \equiv t - t'$ in Eq. (17a) results in the expression

$$[X(t) - iY(t)]e^{i\phi(t)} = -i\int_{0}^{\infty}[\mu\mathcal{E}(t-x)e^{i\phi(t-x)}/\hbar]Z(t-x) \times \exp-(1/T_2' + i\Delta)x\,dx. \tag{18}$$

The form of the integrand above suggests that when the pulse amplitude and phase are slowly varying compared with $\exp-(1/T_2' + i\Delta)x$, it would be useful to use the Taylor expansion

$$\frac{\mu\mathcal{E}(t-x)e^{i\phi(t-x)}Z(t-x)}{\hbar} = \sum_{n=0}^{\infty}\frac{(-)^n x^n}{n!} \times \frac{d^n}{dt^n}\frac{\mu\mathcal{E}(t)e^{i\phi(t)}Z(t)}{\hbar}. \tag{19}$$

When this expression is substituted into Eq. (18) the integral over x can be carried out explicitly yielding

$$[X(t) - iY(t)]e^{i\phi(t)} = i\sum_{n=0}^{\infty}\frac{(-)^{n+1}}{(1/T_2' + i\Delta)^{n+1}} \times \frac{d^n}{dt^n}\left(\frac{\mu\mathcal{E}(t)e^{i\phi(t)}}{\hbar}Z(t)\right). \tag{20}$$

The expansion given by Eq. (20) is the basic formula which will be used to obtain approximate expressions for the response of the atom to an off-resonant light pulse. Intuition suggests that the series converges rapidly when the quantity $(\mathcal{E}e^{i\phi}Z)$ does not vary a great deal in a time $[(1/T_2')^2+\Delta^2]^{-1/2}$.

FIRST-ORDER APPROXIMATION

An alternate derivation of the result shown in Eq. (20) can be obtained by repeatedly integrating Eq. (17a) by parts. A first integration by parts yields the *exact* expressions

$$(X-iY)e^{i\phi}=\frac{-i}{1/T_2'+i\Delta}\,\frac{\mu\mathcal{E}(t)e^{i\phi(t)}}{\hbar}Z(t)+R_1(t)\,, \tag{21a}$$

where

$$R_1(t)=\frac{+i}{1/T_2'+i\Delta}\int_{-\infty}^{t}\frac{d}{dt'}\left(\frac{\mu\mathcal{E}(t')e^{i\phi(t')}Z(t')}{\hbar}\right)\times e^{-(1/T_2'+i\Delta)(t-t')}dt'\,. \tag{21b}$$

When the remainder term is negligible Eq. (21a) can be used to substitute for Y in Eq. (9b). The result is the well-known[10] rate equation

$$\dot{Z}=-\frac{T_2'}{1+(T_2'\Delta)^2}\left(\frac{M\mathcal{E}}{\hbar}\right)^2Z-\frac{(Z-Z_{\text{eq}})}{T_1}\,. \tag{22}$$

The rate equation is usually written in terms of the expectation of an atom's energy which is related to Z according to Eq. (4b).

Also contained in this first-order approximation is the expression for the nonlinear index of refraction which was first derived by Javan and Kelley.[11] To verify this, note that when the longitudinal relaxation time T_1 is short then Eq. (17b) can be expanded in a manner similar to Eq. (17a) to form the series

$$Z(t)=Z(-\infty)+\sum_{n=0}^{\infty}(-T_1)^{n+1}\frac{d^n}{dt^n}\left(\frac{\mu\mathcal{E}(t)Y(t)}{\hbar}\right)\,. \tag{23}$$

When the lifetime T_1 is short compared with the rate of change of the pulse envelope and $(\hbar/\mu\mathcal{E})$ and $(1/\Delta)$, as it is for the systems analyzed in Ref. 11, only the first term in the series of Eq. (23) need be considered. Substituting this approximate expression for Z into Eq. (21a) yields

$$X-iY=\frac{-(i+\Delta T_2')T_2'Z(-\infty)\mathcal{E}}{1+T_1T_2'(\mu\mathcal{E}/\hbar)^2+(\Delta T_2')^2}\,. \tag{24}$$

Substituting this result into Eq. (5) results in a dipole moment per unit volume,

$$\vec{\mathrm{P}}(z,t)=\mathrm{Re}\{(e_x+ie_y)\chi\mathcal{E}\exp[i\phi-i\omega(t-z/c)]\}\,, \tag{25}$$

where the nonlinear susceptibility is

$$\chi=\frac{-N\mu^2}{\hbar}\,\frac{T_2'(i+\Delta T_2')Z(-\infty)}{[1+T_1T_2'(\mu\mathcal{E}/\hbar)^2+(\Delta T_2')^2]}\,. \tag{26}$$

This expression is equivalent to the nonlinear index of refraction derived by Javan and Kelley for a homogeneously broadened system. It should be emphasized that going from Eqs. (21a) and (23) to Eq. (24) required a longitudinal relaxation time T_1 so short that only the $n=0$ term in the expansion of Eq. (23) is significant. Thus the Javan and Kelley solution of Eq. (24) is valid when the pulse envelope varies slowly compared with the relaxation time T_1. The adiabatic-following solution requires the *opposite* condition, i.e., that the pulse be short compared with T_1.

Integrating Eq. (17a) by parts a second time gives the following *exact* expression

$$(X-iY)e^{i\phi}=\frac{-i}{1/T_2'+i\Delta}\,\frac{\mu\mathcal{E}(t)e^{i\phi(t)}}{\hbar}Z(t)+\frac{i}{(1/T_2'+i\Delta)^2}\,\frac{d}{dt}\left(\frac{\mu\mathcal{E}e^{i\phi}Z}{\hbar}\right)+R_2(t)\,, \tag{27a}$$

where

$$R_2(t)=\frac{-i}{(1/T_2'+i\Delta)^2}\int_{-\infty}^{t}\frac{d^2}{dt^2}\left(\frac{\mu\mathcal{E}e^{i\phi}Z}{\hbar}\right)\times\exp\left[-\left(\frac{1}{T_2'}+i\Delta\right)(t-t')\right]dt'\,. \tag{27b}$$

First it will be shown that the adiabatic-following approximation is contained in Eq. (27a) when R_2 is negligible. To do this, consider a pulse without frequency modulation ($\phi=0$) and of a duration short compared with T_1 and T_2'. Under these restrictions the second term of Eq. (27a) may be expanded with the aid of Eq. (9b) as follows:

$$\frac{-i}{\Delta^2}\frac{d}{dt}\left(\frac{\mu\mathcal{E}}{\hbar}Z\right)=\frac{-i}{\Delta^2}Z\frac{d}{dt}\left(\frac{\mu\mathcal{E}}{\hbar}\right)+\frac{i}{\Delta^2}\left(\frac{\mu\mathcal{E}}{\hbar}\right)^2Y\,. \tag{28}$$

When $R_2(t)$ is negligible, the substitution of this expression into Eq. (27a) yields expressions for X and Y in terms of Z,

$$X=-Z(\mu\mathcal{E}/\hbar\Delta)\,, \tag{29a}$$

$$Y=\frac{Z[d(\mu\mathcal{E}/\hbar)/dt]}{(\mu\mathcal{E}/\hbar)^2+\Delta^2}\,. \tag{29b}$$

It follows from Eq. (29) that the ratio,

$$\frac{Y}{Z}=\frac{d(M\mathcal{E}/\hbar)/dt}{(\mu\mathcal{E}/\hbar)^2+\Delta^2},$$

is small compared with unity when the rate of change of the pulse envelope $d(M\mathcal{E}/\hbar)/dt$ is small compared with either Δ^2 or $(\mu\mathcal{E}/\hbar)^2$. The expressions of Eq. (29) can be used in Eq. (8) and the resulting equation solved for Z. From this expression for Z it follows that:

$$X=-\frac{\mu\mathcal{E}}{\hbar}\left(\frac{\Delta}{|\Delta|}\right)\frac{1}{[(\mu\mathcal{E}/\hbar)^2+\Delta^2]^{1/2}}Z(-\infty), \tag{30a}$$

$$Y=\frac{d}{dt}\left(\frac{\mu\mathcal{E}}{\hbar}\right)\frac{|\Delta|}{[(\mu\mathcal{E}/\hbar)^2+\Delta^2]^{3/2}}Z(-\infty), \tag{30b}$$

$$Z=\frac{|\Delta|}{[(\mu\mathcal{E}/\hbar)^2+\Delta^2]^{1/2}}Z(-\infty). \tag{30c}$$

The fact that Y^2 is negligible compared with Z^2 was used in the derivation of Eq. (30).

It is seen that Eqs. (30a)–(30c) are the same as Eqs. (16a)–(16c) which were derived geometrically from the vector model. In addition, the analytic derivation of the adiabatic-following approximation has provided a closed-form expression for the error incurred in making the approximation [Eq. (27b)].

COMPARISON WITH SIT SOLUTION

An exact solution of Eqs. (9a) and (9b) with $T_2'=T_1=\infty$ and $\dot{\phi}=0$ was presented in Ref. 1. This self-induced transparency (SIT) solution can be written

$$\mathcal{E}=\mathcal{E}_0\,\mathrm{sech}(t/\tau), \tag{31a}$$

$$X=\Delta(\mu\mathcal{E}/\hbar)[(1/\tau)^2+\Delta^2]^{-1}, \tag{31b}$$

$$Y=-\frac{d}{dt}\left(\frac{\mu\mathcal{E}}{\hbar}\right)\frac{1}{(1/\tau)^2+\Delta^2}, \tag{31c}$$

$$Z=-\frac{[(\mu\mathcal{E}_0/2\hbar)^2+\Delta^2]-\frac{1}{2}(\mu\mathcal{E}/\hbar)^2}{(1/\tau)^2+\Delta^2}, \tag{31d}$$

for the initial condition $Z(-\infty)=-1$. The duration τ and amplitude $\mathcal{E}_0$ of the SIT pulse are related according to

$$\tau(\mu\mathcal{E}_0/\hbar)=2. \tag{32}$$

It is easy to show that the exact SIT solution given in Eq. (31) reduces to the approximate adiabatic-following solution of Eq. (30) when the square of the pulse duration τ is much larger than the square of the precession time $1/\Delta$. In view of the condition of Eq. (32), this restricts both the duration and amplitude of the light pulse as follows:

$$\Delta^2\gg(1/\tau)^2 \tag{33a}$$

and

$$\Delta^2\gg(\mu\mathcal{E}/\hbar)^2. \tag{33b}$$

It follows from Eq. (33b) that the special connection between the pulse duration and amplitude of an SIT pulse restricts the region of overlap with the adiabatic-following solution to the linear regime.[12]

ESTIMATE OF ERROR

An estimate of the error incurred in using the adiabatic-following approximation given by Eqs. (30) will be made in this section. To accomplish this, substitute Eq. (30c) into the expression for the error term which is shown in Eq. (27b). In the limit that $1/T_2'$ is negligible, it is seen that the error term is given by

$$R_2(t)e^{i\Delta t}=\frac{i}{\Delta^2}\int_{-\infty}^{t}\frac{|\Delta|}{\sqrt{\epsilon^2+\Delta^2}}\left(\frac{d^2\epsilon}{dt^2}-\frac{3\epsilon(d\epsilon/dt)^2}{\epsilon^2+\Delta^2}\right)\times e^{i\Delta t}dt Z(-\infty), \tag{34}$$

where the short-hand notation

$$\epsilon=\mu\mathcal{E}/\hbar \tag{35}$$

has been introduced. An investigation of the integral in Eq. (34) for a particular pulse shape would in general require a numerical integration. An analytic expression can be found, however, for a pulse envelope which is given by

$$\epsilon=\epsilon_0(t^2/\tau^2)e^{-t/\tau}U(t), \tag{36}$$

where $U(t)$ is the unit step function, and if it is assumed that the inequality

$$\Delta^2\gg\epsilon^2 \tag{37}$$

is satisfied. A graph of the pulse envelope defined by Eq. (36) is shown in Fig. 2. Comparison with the adiabatic-following expression for Z, given in Eq. (30c), reveals that the condition $\Delta^2\gg\epsilon^2$ implies that $Z(z,t,\Delta)\simeq\pm1$, and the atomic system's response is linear.[13] When Eq. (36) is substituted into Eq. (34) under the restriction of Eq. (37) the resulting expression for the error term is given by

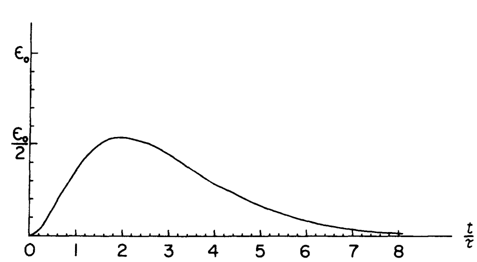

FIG. 2. Pulse envelope of Eq. (36) is shown in this graph.

$$R_2 = \frac{-i\epsilon_0}{\Delta^2\tau(1-i\Delta\tau)}\left[\frac{2(\Delta\tau)^2}{(1-i\Delta\tau)^2}e^{-i\Delta t} + e^{-t/\tau}\left(\frac{t^2}{\tau^2} - \frac{2(1-2i\Delta\tau)}{(1-i\Delta\tau)}\frac{t}{\tau} - \frac{2(\Delta\tau)^2}{(1-i\Delta\tau)^2}\right)\right]Z(-\infty). \quad (38)$$

If relaxation processes had been included, the first term within the brackets of Eq. (38) would show a damped oscillating time dependence of the form $e^{-(1/T_2'+i\Delta)t}$. This transient term occurs because the light pulse is turned on at a definite time. It is seen that the error term in Eq. (38) is of order $R_2 \sim \epsilon_0/\Delta^3\tau^2$ for large $\Delta\tau$. The adiabatic-following expressions for X and Y, which correspond to the light pulse of the form of Eq. (36), are

$$X = -(\epsilon_0/\Delta)(t^2/\tau^2)e^{-t/\tau}U(t)Z(-\infty) \quad (39a)$$

and

$$Y = (\epsilon_0/\Delta^2\tau)[(2t/\tau) - (t^2/\tau^2)]e^{-t/\tau}U(t)Z(-\infty), \quad (39b)$$

respectively. Comparing Eqs. (38) and (39a) and (39b), it is seen that the ratios of the error term to X and Y are of the order of

$$R_2/X \sim 1/(\Delta\tau)^2 \quad (40a)$$

and

$$R_2/Y \sim 1/(\Delta\tau) \quad (40b)$$

for large values of $\Delta\tau$. It is thus seen that in the limit of Eq. (37), the adiabatic-following expression will be a good approximation for X when $(\Delta\tau)^2 \gg 1$ is satisfied and a good approximation for Y when $(\Delta\tau) \gg 1$ is satisfied.

The adiabatic-following approximation is not limited to the region described by Eq. (37). To see that it has validity in the nonlinear region (where $\mu\mathcal{E}/\hbar\Delta$ is not negligible), the expression for the error term given in Eq. (34) was investigated numerically for a light pulse which has a Gaussian time dependence,

$$\epsilon(t) = \epsilon_0 e^{-t^2/2\tau^2}. \quad (41)$$

For this pulse shape the error term is given by

$$R_2(t)e^{i\Delta t} = [(-i\epsilon_0)/\Delta^2\tau]I(t,\epsilon_0,\Delta)Z(-\infty), \quad (42a)$$

where

$$I(t,\epsilon_0,\Delta) = \int_{-\infty}^{t/\tau}\left((u^2-1)e^{-u^2/2}\frac{-3(\epsilon_0/\Delta)^2u^2e^{-3u^2/2}}{1+(\epsilon_0/\Delta)^2e^{-u^2}}\right) \times \frac{e^{i\Delta\tau u}}{[1+(\epsilon_0/\Delta)^2e^{-u^2}]^{1/2}}\,du. \quad (42b)$$

The corresponding adiabatic-following expressions for X and Y are

$$X = -\frac{\epsilon_0}{\Delta}\frac{e^{-t^2/2\tau^2}}{[1+(\epsilon_0/\Delta)^2e^{-t^2/\tau^2}]^{1/2}}Z(-\infty) \quad (43a)$$

and

$$Y = -\frac{\epsilon_0}{\Delta^2\tau}\frac{(t/\tau)e^{-t^2/2\tau^2}}{[1+(\epsilon_0/\Delta)^2e^{-t^2/\tau^2}]^{3/2}}Z(-\infty), \quad (43b)$$

respectively. As long as the amount off-resonance is sufficiently large that $\epsilon_0 \lesssim 4\Delta$, a comparison of Eqs. (42a) and (43a) with (43b) reveals that the fractional error incurred by using the adiabatic-following expressions will be

$$R_2/X \sim I(t,\epsilon_0,\Delta)/(\Delta\tau) \quad (44a)$$

and

$$R_2/Y \sim I(t,\epsilon_0,\Delta) \quad (44b)$$

for the variables X and Y. It can be expected that for $\Delta\tau$ sufficiently large the oscillating exponential in Eq. (42b) will lead to cancellation that will decrease the value of the integral.

For constant t and Δ, the integral of Eq. (42b) drops off as $1/\epsilon_0$ when the pulse amplitude ϵ_0 becomes large. This result suggests that the conclusions of Eqs. (40), which were reached in the linear limit, will not break down in the nonlinear limit.

The integral $I(t,\epsilon_0,\Delta)$ has been studied numerically for a wide range of values of t, ϵ_0/Δ, and $\Delta\tau$. Figure 3 shows a three-dimensional graph of the absolute values of I as a function of ϵ_0/Δ and $\Delta\tau$ for the particular times $t=0$ and $0.5\ \tau$. A study of this and other plots indicates that $I(t,\epsilon_0,\Delta)$ drops off at least as fast as $1/\Delta\tau$ for large $\Delta\tau$ and values of pulse amplitude $\epsilon/\Delta = \mu\mathcal{E}_0/\hbar\Delta \lesssim 4$. According to Eqs. (44), this result indicates that the adiabatic-following expression for X will be valid when $(\Delta\tau)^2 \gg 1$ and the expression for Y will be valid when $(\Delta\tau) \gg 1$. This conclusion holds for pulse amplitudes $\epsilon_0 = (\mu\mathcal{E}_0/\hbar)$ that are less than or comparable to the amount off-resonance 4Δ.

The conclusion of this section can be expressed in physical terms as follows. The adiabatic-following approximation will be valid when the product of pulse duration τ times the precessional frequency Δ is large compared with one. Alternately it is required that the pulse bandwidth $1/\tau$ be much less than the amount off-resonance Δ. The two statements are essentially equivalent for a light pulse with a smooth envelope.

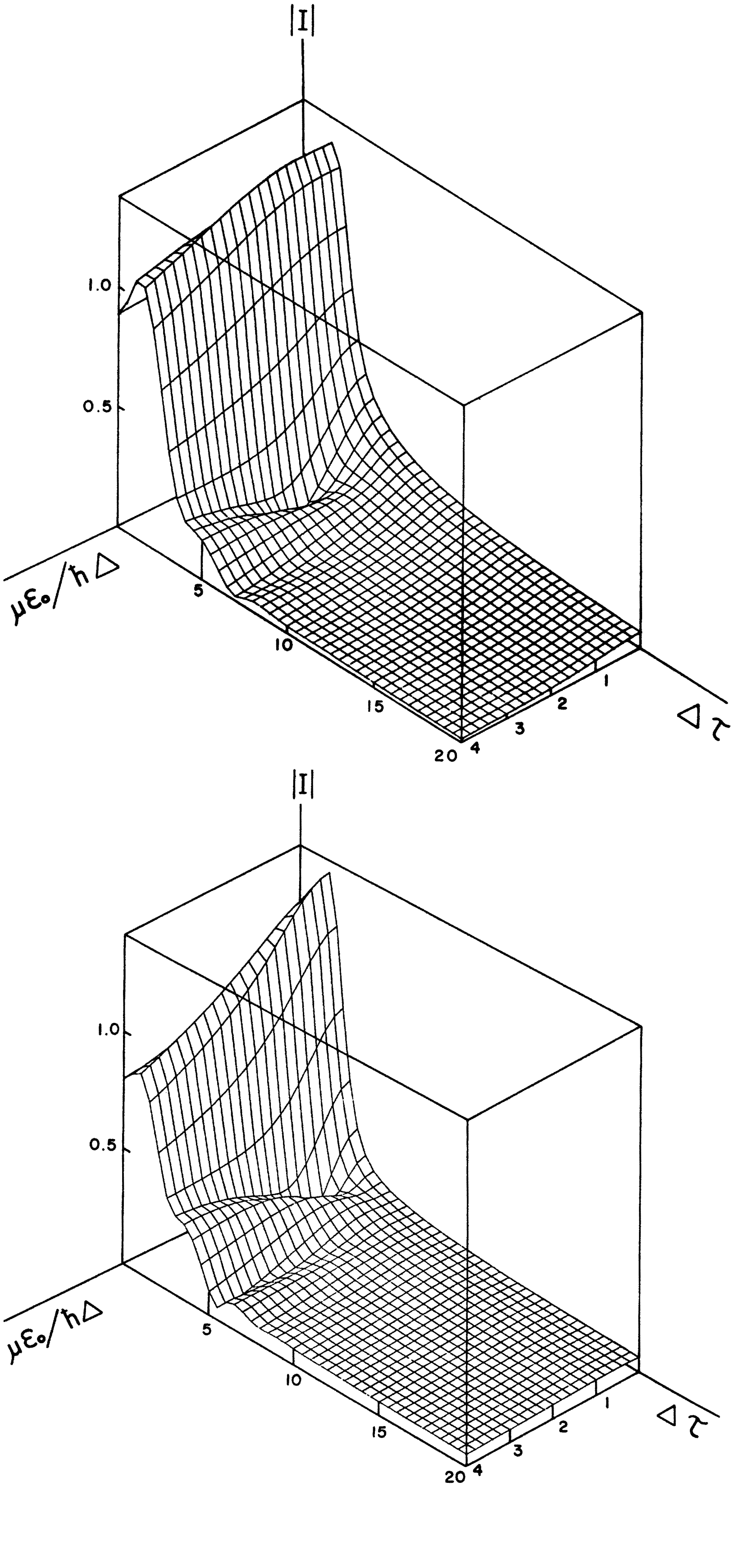

FIG. 3. (a) Three-dimensional plot of the absolute value of the integral of Eq. (42b) as a function of pulse amplitude $\epsilon_0=\mu\mathcal{E}_0/\hbar$ and amount off resonance $\Delta=\Omega-\omega$ is shown in this figure. The graph corresponds to the particular time $t=0$. (b) Graph of $|I|$ for the particular time $t=0.5\tau$.

PROPAGATIONAL EFFECTS

In the adiabatic limit, the polarization of a resonant medium can be found by substituting Eqs. (30a) and (30b) into Eq. (5). When the amount off resonance Δ is large compared with the inhomogeneous linewidth $1/T_2^*$, the integral involving the line-shape function $g(\Delta)$ can be factored out and set equal to unity. Thus, the polarization is then

$$\vec{\mathrm{P}}(z,t)=-\frac{N\mu^2}{\hbar}\,\mathrm{Re}\left\{(\hat{e}_x+i\hat{e}_y)\left[\frac{\Delta}{|\Delta|}\frac{1}{[(\mu\mathcal{E}/\hbar)^2+\Delta^2]^{1/2}}\,\mathcal{E}+i\frac{d\mathcal{E}}{dt}\frac{|\Delta|}{[(\mu\mathcal{E}/\hbar)^2+\Delta^2]^{3/2}}\right]\exp\{-i[\omega(t-z/c)-\phi]\}\right\}Z(-\infty)\,. \tag{45}$$

For large $\Delta\tau$ (where τ is the pulse duration), the term containing $d\mathcal{E}/dt$ in Eq. (45) will be small compared with the first term. When this term is neglected, Eq. (45) can be written in the form of Eq. (25) where the nonlinear susceptibility is equal to

$$\chi=-\frac{N\mu^2}{\hbar}\frac{\Delta}{|\Delta|}\frac{Z(-\infty)}{[(\mu\mathcal{E}/\hbar)^2+\Delta^2]^{1/2}}\,. \tag{46}$$

A nonlinear susceptibility of this form can give rise to self-focusing or self-defocusing. Experimental observation of these two effects together with their analysis can be found in Refs. 3 and 4. Note that the nonlinear susceptibility that follows from the adiabatic-following approximation is of a different functional form than Javan and Kelley's result shown in Eq. (26).

Propagation of an off-resonant light pulse in the adiabatic limit is described by substituting Eqs. (30a) and (30b) into Eq. (10). Evolution of the amplitude $\epsilon=\mu\mathcal{E}/\hbar$ and phase ϕ of the light pulse is then described by two simultaneous nonlinear differential equations,

$$\frac{\partial\epsilon}{\partial z}+\left(\frac{1}{c}+\frac{\alpha_0|\Delta|(2\Omega-\omega-2\dot{\phi})}{\omega(\epsilon^2+\Delta^2)^{3/2}}\right)\frac{\partial\epsilon}{\partial t}=0\,, \tag{47a}$$

$$\frac{\partial\phi}{\partial z}+\frac{1}{c}\frac{\partial\phi}{\partial t}=+\frac{\alpha_0(\omega-2\dot{\phi})\Delta}{\omega|\Delta|(\epsilon^2+\Delta^2)^{1/2}}\,, \tag{47b}$$

where the parameter

$$\alpha_0\equiv-(2\pi N\mu^2/\hbar c)\omega Z(-\infty) \tag{48}$$

has been introduced. This definition of α_0 gives a positive number when the atoms are prepared in their ground state. For most practical experiments the carrier frequency of the light pulse ω will be much larger than the amount of off-resonance Δ and frequency modulation $\dot{\phi}$. Thus it is a good approximation to replace these equations by the decoupled equations,

$$\frac{\partial\epsilon}{\partial z}+\left[\frac{1}{c}+\frac{\alpha_0|\Delta|}{(\epsilon^2+\Delta^2)^{3/2}}\right]\frac{\partial\epsilon}{\partial t}=0\,, \tag{49a}$$

$$\frac{\partial\phi}{\partial z}+\frac{1}{c}\frac{\partial\phi}{\partial t}=+\frac{\alpha_0\Delta}{|\Delta|(\epsilon^2+\Delta^2)^{1/2}}\,. \tag{49b}$$

Equation (49a) is satisfied by a solution of the form[5,14]

$$\epsilon=F(t-z/v)\,, \tag{50a}$$

where the intensity-dependent group velocity is given by

$$v^{-1}=c^{-1}+[\alpha_0|\Delta|/(\epsilon^2+\Delta^2)^{3/2}]\,. \tag{50b}$$

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