Coherent two-photon resonance and Doppler-free population inversion*

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(Received 15 March 1976)

Doppler-free population inversion induced by coherent two-photon resonance is calculated analytically for a two-photon analog of the hyperbolic-secant pulse and numerically for a Gaussian, linearly chirped pulse. The numerical calculations are carried out for a two-photon resonance of a chirped ruby laser line with the $6S_{1/2} \rightarrow 9D_{3/2}$ transition in Cs. Complete inversion is found when the fractional change in frequency during the pulse lies between 6×10^{-6} and 2×10^{-5}. The coherent two-photon resonances are described by optical Bloch equations which are derived from a multiple-time-scale perturbation theory.

I. INTRODUCTION

The inversion of level populations by a one-photon coherent resonance is a familiar idea, especially in the context of the adiabatic-following approximation.[1] In this connection Grischkowsky, Loy, and Liao[2] have recently described an extension of adiabatic following to the case of two-photon resonances, and Grischkowsky and Loy[3] have pointed out the possibility of self-induced adiabatic rapid passage which is unique to the two-photon case. In the present paper two-photon induced population inversions are considered in the special case of counterpropagating beams. This provides the well-known[3,4] advantage of reducing or eliminating Doppler broadening. This feature has possible applications in spectroscopy,[4] selective control of chemical reactions,[5-7] and isotope separation.[8]

In this paper the theory of the two-photon coherent resonance is derived from the multiple-time-scale (MTS) perturbation theory described in a previous publication[9] (hereafter referred to as WGE); the terminology and notation of WGE will be used here. The essential assumption of the WGE method is that there are several widely separated time scales in the problem. In the present application there are two basic time scales: the fast scale defined by the laser oscillation period and the slow scale given by the Rabi oscillation period of the target system. The small parameter of the theory is the ratio λ of the Rabi frequency to the laser frequency; it represents the dipole coupling strength. The formalism yields the wave function as an expansion in λ with coefficients whose time dependence also involves λ. This provides a systematic means for extracting the coherent effects of single- and many-photon resonances that are not correctly treated by conventional perturbation theory. It was shown in WGE that this expansion satisfies conservation of probability (unitarity) order by order in λ.

According to the WGE method the conditions for a two-photon coherent resonance are that the fractional pulse width and fractional detuning, near the pulse maximum, should both be of order λ^2; the more stringent conditions of the adiabatic-following approximation are not required. In the absence of one-photon resonances, the leading approximation replaces the many-level system by an effective two-level system with an interaction Hamiltonian whose matrix elements include the effects of nonresonant levels. An added advantage of the WGE method is that it can also be used in the presence of resonances with intermediate states. Examples of such use will be presented elsewhere.

The general problem of developing approximate theories for situations involving widely separated time scales has been approached in many different ways for applications in planetary orbit theory,[10] nonlinear mechanics,[11] nonlinear optics,[12(a)] fluid mechanics,[12(b)] and the derivation of kinetic equations.[13,14] The WGE method is an adaptation of a technique employed in deriving kinetic equations from the hierarchy of equations for reduced distribution functions in statistical mechanics. In applications to statistical mechanics and quantum mechanics, two difficulties must be surmounted: the presence of secular terms (unbounded as $t \rightarrow \infty$) and quasisecular terms (containing small energy denominators). The standard references on the multiple-scales method[15] show how to avoid secular but not quasisecular terms. A satisfactory solution of the quasisecularity problem in statistical mechanics is given by Sandri,[13] and a somewhat different technique is used in the WGE treatment of the Schrödinger equation.[9]

The special problem of the two-photon resonance has also been approached in several ways.[16-20] The theories of Belenov and Poluektov[16] and Takatsuji[17] reduce a general multilevel system to an effective two-level system consisting of the resonantly coupled levels. This result requires two assumptions: (1) All intermediate levels are far off resonance. (2) The fields are adiabatic with respect to the intermediate states; i.e., $|\partial \mathcal{E}/\partial t| \ll |\mathcal{E}| \Delta\omega$, where $\Delta\omega$ is the minimum detuning for the intermediate states. These conditions are not required by the general WGE method; however, this makes no practical difference for the present paper since the specific application presented does satisfy both conditions. Thus, the effective Bloch equations used in Sec. III may be derived from the WGE theory, as in Sec. II, or from the Takatsuji theory, as in the work of Grischkowsky, Loy, and Liao.[2]

A different approach is found in the work of Brewer and Hahn[19] and Beterov and Chebotaev.[20] These authors discuss three-level systems irradiated by two laser beams, each being approximately resonant with one of the transitions joining the intermediate state to the initial and final states. In certain cases exact solutions (of the rotating-wave approximation) were found.

The same equations are obtained in the lowest approximation of the WGE method; in that language they would be said to describe coupled one-photon transitions. This distinction is more than a matter of convention since the solution of this problem exhibits oscillations with frequencies $\Omega \sim O(\lambda)\bar{\omega}$, where $\bar{\omega}$ characterizes the laser fields; e.g., $\bar{\omega}$ might be the average of the input frequencies. On the other hand, the strictly two-photon transitions, for which the intermediate states are virtual, yield oscillation frequencies $\omega \sim O(\lambda^2)\bar{\omega}$. The next approximation in the WGE method gives a modulation of the coupled one-photon solution by frequencies of this order.

In Sec. II the relevant results from WGE are used to obtain the effective two-level Hamiltonian in the absence of resonances with intermediate levels. In Sec. III the corresponding optical Bloch equations are presented for the case of counter-propagating beams. An exact analytical solution for the effective two-level system is constructed from the known solution for the hyperbolic-secant pulse in the one-photon problem. Some features of the chirped-pulse technique which are peculiar to the two-photon case are pointed out.

In Sec. IV the theory of Sec. III is applied to a hypothetical experiment in which a ruby laser is chirped through the two-photon resonance frequency for the transition $6S_{1/2} \to 9D_{3/2}$ in Cs. For given values of pulse intensity and duration, essentially complete inversion is attained for a range of values of the chirp rate. These pulses are roughly adiabatic, but the solutions clearly deviate from the small-angle adiabatic-following model.

II. TWO-PHOTON COHERENT RESONANCE

The target atom or molecule will be represented by an N-level system with energies E_α, eigenfunctions ϕ_α, and interaction Hamiltonian $H_1 = -\vec{\mathcal{E}}(t)\cdot\vec{d}$, where $\vec{d}$ is the dipole operator. Let $\vec{\mathcal{E}} = \vec{\mathcal{E}}_1 + \vec{\mathcal{E}}_2$, where

$$\vec{\mathcal{E}}_k(t) = \tfrac{1}{2}\vec{\mathcal{E}}_{k0}[G_k(t)e^{-i\omega_k t} + \text{c.c.}],$$

$G_k(0) = 1$, and the Fourier transform $\hat{G}_k(\omega)$ is peaked at $\omega = 0$. The width $\delta\omega_k$ of $\hat{G}_k$ must satisfy $\delta\omega_k \leq O(\lambda)\bar{\omega}$, where $\bar{\omega}$ is the frequency characterizing the laser; otherwise the pulse duration $\tau_k \sim 1/\delta\omega_k$ would satisfy $\tau_k \ll (\lambda\bar{\omega})^{-1}$ and the pulse would be over before anything could happen on the time scale of the Rabi period $(\lambda\bar{\omega})^{-1}$. The interaction-picture Schrödinger equation is ($\hbar = 1$)

$$i\,\frac{\partial\Psi}{\partial t} = \lambda H^{(1)}(t)\Psi, \quad H^{(1)}(t) = e^{itH_0}H_1(t)e^{-itH_0}, \tag{2.1}$$

where H_0 is the unperturbed Hamiltonian and λ is a dimensionless parameter representing the dipole coupling strength. For estimation purposes λ is defined by $\lambda = \mathcal{E}_0\bar{d}/\bar{\omega}$, where $\mathcal{E}_0 = \max|\vec{\mathcal{E}}(t)|$, $\bar{d}$ is a representative value for the dipole matrix elements and $\bar{\omega}$ is the characteristic frequency for $\vec{\mathcal{E}}(t)$. With this definition of λ, H_1 should be replaced by

$$H_1' = -(\bar{\omega}/\mathcal{E}_0\bar{d})\,\vec{\mathcal{E}}\cdot\vec{d},$$

but this distinction will be ignored since, at the end of the calculation, the factors λ and H_1' can always be recombined to recover H_1.

The multiple-time-scale method involves the extensions

$$\begin{aligned} &t \to (t_0, t_1, t_2, \ldots), \\ &\frac{\partial}{\partial t} \to \sum_{n=0}^{\infty} \lambda^n \frac{\partial}{\partial t_n}, \\ &\Psi \to \sum_{n=0}^{\infty} \lambda^n \Psi^{(n)}(t_0, t_1, \ldots). \end{aligned} \tag{2.2}$$

The extended time derivative is the directional derivative along the line $t_n = \lambda^n t$ (the physical line); at the end of the calculation an approximate solution of (2.1) is obtained by evaluating $\Psi^{(n)}$ on the physical line. An infinite set of equations for the $\Psi^{(n)}$'s results from substituting the extended forms (2.2) into (2.1) and equating equal *explicit* powers of λ on both sides. These equations are not sufficient to determine the $\Psi^{(n)}$'s; furthermore, straightforward integration yields two kinds of troublesome terms: (1) secular terms, proportional to some t_k and (2) quasisecular terms, containing energy denominators of order λ. In Sec. II of WGE a general prescription was given for imposing subsidiary conditions which eliminate the secular and quasisecular terms and also allow the $\Psi^{(n)}$'s to be completely determined. The results of this prescription applied to the present problem are given below.

The lowest-order approximation in which resonant two-photon effects can occur requires the evaluation of $\Psi^{(0)}$ as a function of t_0, t_1, and t_2. The $\Psi^{(n)}$ $(n \geq 1)$ and the dependence of $\Psi^{(0)}$ on $t_3, t_4, \ldots$ can be neglected. The explicit λ on the right-hand side of (2.1) guarantees that $\Psi^{(0)}$ is independent of t_0, but the t_1 and t_2 dependences must be obtained from the auxiliary conditions which eliminate t_0 and t_1 secularities. The first of these is given by Eq. (2.11) of WGE:

$$i\frac{\partial}{\partial t_1}\Psi^{(0)} = H^{(1;1)}\Psi^{(0)}\,, \quad H^{(1;1)}(t_1) = \mathcal{P}H^{(1)}(\lambda^{-1}t_1)\,.$$

In second equation $\mathcal{P}$ is a projection operator that selects the static and low-frequency parts of its operand. The exact definition is

$$\mathcal{P}A(t_k) = \langle A\rangle_k + \int\frac{d\omega}{2\pi}\theta(\lambda^2\bar{\omega}^2 - \omega^2)[\hat{A}'(\omega)]_0 e^{-i\omega t_k}\,,$$

where $A' = A - \langle A\rangle_k$, $\bar{\omega}$ is a suitable frequency unit; e.g., the average frequency of the pulse, a circumflex (^) denotes the Fourier transform, $[B]_0$ is an instruction to delete any part of B which is of order λ or smaller, and

$$\langle A\rangle_k \equiv \lim_{T\to\infty}\frac{1}{T}\int_0^T dt_k' A(t_k')$$

is the static part of A. The parts of A that appear in $\mathcal{P}A$ are precisely those that would lead to secularities and quasisecularities in conventional perturbation theory. The matrix elements of $H^{(1)}$ are

$$H^{(1)}_{\alpha\beta} = \sum_{k=1}^{2}\tfrac{1}{2}(V_k)_{\alpha\beta}e^{itE_{\alpha\beta}}[G_k(t)e^{-i\omega_k t} + \text{c.c.}], \qquad (2.3)$$

where $E_{\alpha\beta} \equiv E_\alpha - E_\beta$ and $(V_k)_{\alpha\beta} = -\vec{\mathcal{E}}_{k0}\cdot\vec{d}_{\alpha\beta}$. With the help of the condition $\delta\omega_k \lesssim O(\lambda)\bar{\omega}$, it is not difficult to show that $\mathcal{P}H^{(1)}_{\alpha\beta} = 0$ unless $|E_{\alpha\beta} \pm \omega_k| = O(\lambda)\bar{\omega}$. In other words $H^{(1;1)}$ vanishes unless one of the fields is in resonance with some transition in the target.[21] The general formalism in WGE can deal with the simultaneous presence of one- and two-photon resonances, but the example considered in Sec. IV involves no one-photon resonances; therefore, the condition $|E_{\alpha\beta} \pm \omega_k| \gg O(\lambda)\bar{\omega}$ will be assumed from here on.

The absence of one-photon resonances considerably simplifies the auxiliary condition determining the t_2 dependence of $\Psi^{(0)}$. The condition is given by Eq. (2.25) of WGE:

$$i\frac{\partial}{\partial t_2}\Psi^{(0)} = H^{(2;2)}\Psi^{(0)},$$

$$H^{(2;2)}(t_2) = \mathcal{P}X^{(2)}(\lambda^{-1}t_2), \qquad (2.4)$$

$$X^{(2)}(t_1) = \mathcal{P}[H^{(1)}J^{(1;0)}](\lambda^{-1}t_1), \qquad (2.5)$$

$$J^{(1;0)}(t_0) = \text{P}\int\frac{d\omega}{2\pi}\frac{\hat{H}^{(1)}(\omega)}{\omega}e^{-i\omega t_0}, \qquad (2.6)$$

where P stands for principal value. Substitution of (2.3) into (2.5) and (2.6) yields

$$X^{(2)}_{\alpha\beta}(t_1) = \mathcal{P}\sum_k\sum_j\sum_\gamma \text{P}\int\frac{d\omega}{2\pi}\int\frac{d\omega'}{2\pi}\exp[-i(\omega/\lambda)t_1]\theta(\lambda^2\bar{\omega}^2 - \omega^2)\frac{1}{8}\left(\frac{(V_k)_{\alpha\gamma}(V_j)_{\gamma\beta}}{\omega'} - \frac{(V_j)_{\alpha\gamma}(V_k)_{\gamma\beta}}{\omega - \omega'}\right)$$
$$\times[\hat{G}_k(\omega - \omega' + E_{\alpha\gamma} - \omega_k) + \hat{G}_k(-\omega + \omega' - E_{\alpha\gamma} - \omega_k)^*]$$
$$\times[\hat{G}_j(\omega' + E_{\gamma\beta} - \omega_j) + \hat{G}_j(-\omega' - E_{\gamma\beta} - \omega_j)^*]. \qquad (2.7)$$

This expression separates into four terms corresponding to the products $\hat{G}_k\hat{G}_j$, etc. Since the $\mathcal{P}$ operation discards any low-frequency term which is $O(\lambda)$ or smaller, a given product can contribute to (2.7) only if the arguments of both $\hat{G}$ functions are small. The combination of this consideration with the restriction $|\omega| \leq \lambda\bar{\omega}$ leads to three conditions for a nonvanishing contribution to (2.7): (a) $|E_{\alpha\beta} - (\omega_1 + \omega_2)| \leq O(\lambda\bar{\omega})$, (b) $|E_{\alpha\beta} - 2\omega_k| \leq O(\lambda\bar{\omega})$, and (c) $|E_{\alpha\beta} - (\omega_k - \omega_j)| \leq O(\lambda\bar{\omega})$. Condition (a) evidently corresponds to absorption (or emission) of one photon from each beam; condition (b) indicates absorption (emission) of two photons from one beam; and condition (c) for $k \neq j$, $\alpha \neq \beta$ represents coherent Raman scattering. Note that (c) is trivially satisfied for $\alpha = \beta$, $k = j$. The counterpropagating problem considered in Sec. III corresponds to (a); therefore it is assumed that only (a) and the trivial version of (c) are satisfied. More specifically, suppose that two states ϕ_s and ϕ_f are connected by a two-photon resonance: $E_{fs} - (\omega_1 + \omega_2) \leq O(\lambda)\bar{\omega}$. Then the only off-diagonal elements of $X^{(2)}$ are $X^{(2)}_{fs} = X^{(2)*}_{sf}$, and (2.7) gives

$$X^{(2)}_{fs} = \int\frac{d\omega}{2\pi}e^{i\omega t_1}\lambda\theta(\lambda^2\bar{\omega}^2 - \lambda^2\omega^2)\int\frac{d\omega'}{2\pi}\sum_\beta\Big[\hat{G}_1(\lambda\omega - \omega' + E_{f\beta} - \omega_1)\hat{G}_2(\omega' + E_{\beta s} - \omega_2)$$
$$\times\frac{1}{8}\left(\frac{(V_1)_{f\beta}(V_2)_{\beta s}}{\omega_2 - E_{\beta s}} - \frac{(V_2)_{f\beta}(V_1)_{\beta s}}{\omega_1 - E_{f\beta}}\right) + (1 \to 2)\Big]. \qquad (2.8)$$

In deriving this expression the denominators ω' and $\omega - \omega'$ have been evaluated at the peak of the product

$\hat{G}_1\hat{G}_2$; this is permitted because the error terms are of order λ or smaller and are eliminated by the $\mathcal{P}$ operation. In addition a change of variables $\omega\to\lambda\omega$ has been made in order to display the t_1 Fourier transform explicitly. Equation (2.4) gives

$$H_{fs}^{(2;2)}=\int\frac{d\omega}{2\pi}\exp[-i(\omega/\lambda)t_2][\theta(\lambda^2\overline{\omega}^2-\omega^2)\hat{X}_{fs}^{(2)}(\omega)]_0. \tag{2.9}$$

It is convenient to change the integration variable $\omega\to\omega/\lambda$ before inserting (2.8) into (2.9) to get

$$H_{fs}^{(2;2)}=\int\frac{d\omega}{2\pi}\exp\left(-i\frac{\omega}{\lambda^2}t_2\right)\left\{\theta(\lambda^4\overline{\omega}^2-\omega^2)\,\theta(\lambda^2\overline{\omega}^2-\omega^2)\int\frac{d\omega'}{2\pi}\sum_\beta\left[\hat{G}_1(\omega-\omega'+E_{f\beta}-\omega_1)\hat{G}_2(\omega'+E_{\beta s}-\omega_2)\times\frac{1}{8}\left(\frac{(V_1)_{f\beta}(V_2)_{\beta s}}{\omega_2-E_{\beta s}}-\frac{(V_2)_{f\beta}(V_1)_{\beta s}}{\omega_1-E_{f\beta}}\right)+(1\to2)\right]\right\}_0. \tag{2.10}$$

In order to get a nonvanishing contribution it is necessary to impose new restrictions: $\delta\omega_k=O(\lambda^2)\overline{\omega}$ and $E_{fs}-(\omega_1+\omega_2)=O(\lambda^2)\overline{\omega}$. If these conditions were violated the ω' integral would be at most $O(\lambda)$ and the $[\cdots]_0$ operation would set the right-hand side of (2.10) to zero. With the new restrictions in force, the constraint $|\omega|\le\lambda^2\overline{\omega}$ on the ω integral can be relaxed; the $\hat{G}$ functions will have the same effect except for small errors which are eliminated by the $[\cdots]_0$ instruction. The frequency integrals can then be performed, by means of the convolution theorem, to get

$$H_{fs}^{(2;2)}=\frac{1}{8}\sum_\beta\left(\frac{(2E_\beta-E_f-E_s)(V_2)_{f\beta}(V_1)_{\beta s}}{(\omega_1-E_{\beta s})(\omega_1-E_{f\beta})}+(1\to2)\right)G_1(\lambda^{-2}t_2)G_2(\lambda^{-2}t_2)\exp\left(i\frac{E_{fs}-\omega_1-\omega_2}{\lambda^2}t_2\right). \tag{2.11}$$

A similar calculation yields the diagonal elements

$$H_{\alpha\alpha}^{(2;2)}=\sum_k\left(\sum_\beta\frac{2E_{\alpha\beta}|(V_k)_{\alpha\beta}|^2}{(E_{\alpha\beta})^2-\omega_k^2}\right)|G_k(\lambda^{-2}t_2)|^2. \tag{2.12}$$

Since $H_{fs}^{(2;2)}$ and $H_{sf}^{(2;2)}$ are the only nonvanishing off-diagonal matrix elements, the N-level system has been effectively replaced by a two-level system described by the Hamiltonian $H_{\alpha\beta}^{(2;2)}$ $(\alpha,\beta=s,f)$. The effects of nonresonant levels are included in the sums over intermediate states in (2.11) and (2.12).

III. COUNTERPROPAGATING BEAMS

Let two pulses propagating in the $\pm z$ directions be described by

$$\vec{\mathcal{E}}_\pm(t)=\vec{\mathcal{E}}_0S(t\mp(1/c)z)\cos[\omega_Lt\mp(1/c)\omega_Lz+\phi(t\mp(1/c)z)],$$

where S and ϕ are respectively the pulse shape and frequency modulation, normalized by $S(0)=1$, $\phi(0)=0$. A target system with velocity $\vec{v}$ experiences the total field $\vec{\mathcal{E}}=\vec{\mathcal{E}}_1+\vec{\mathcal{E}}_2$ with $\vec{\mathcal{E}}_{k0}=\vec{\mathcal{E}}_0$, $\omega_k=(1\pm\beta)\omega_L$, and

$$G_k(t)=S((1\pm\beta)t\pm t_c)\exp[\mp i\omega_Lt_c\pm i\phi((1\pm\beta)t\pm t_c)],$$

where the lower (upper) sign corresponds to $k=1$ $(k=2)$. The parameters β and t_c are given by $\beta=v_3/c$ and $t_c=z_c/c$, where z_c is the location of the target at $t=0$. The maximum overlap of the pulses occurs at $t=0$, $z=0$. Since the shape functions are evaluated at $\pm t_c$ when $t=0$, only targets having $|t_c|\ll\tau$, where τ is the pulse length, will interact strongly with the fields. Thus the active region has a length $L\ll c\tau$. For targets in this region t_c may be neglected in the arguments of S and ϕ; when this is done, it is easy to check that the matrix elements $H_{\alpha\beta}^{(2;2)}$, given in (2.11) and (2.12), are even functions of β. Consequently, the β dependence of the matrix elements can be neglected with an error of order β^2. The detuning $E_{fs}-(\omega_1+\omega_2)=E_{fs}-2\omega_L$ is independent of β and can be set to zero with no loss of generality. In this approximation [neglect of $O(\beta^2)$ terms] all Doppler effects are eliminated from the effective Hamiltonian which is given by

$$H_{fs}^{(2;2)}=\Gamma S^2(t)e^{-2i\phi(t)},$$

$$H_{\alpha\alpha}^{(2;2)}=2\Delta_\alpha S^2(t),$$

$$\Gamma=\frac{1}{4}\sum_\beta\frac{(2E_\beta-E_f-E_s)(V)_{f\beta}(V)_{\beta s}}{(\omega_L-E_{\beta s})(\omega_L-E_{f\beta})}, \tag{3.1}$$

$$\Delta_\alpha=\frac{1}{2}\sum_\beta\frac{E_{\alpha\beta}|(V)_{\alpha\beta}|^2}{(E_{\alpha\beta})^2-\omega_L^2}, \tag{3.2}$$

where t_2 has been evaluated on the physical line $t_2=\lambda^2t$. The quantities Γ and Δ_α are respectively the width of the two-photon resonance and the optical Stark shift of the level α. The absence of Doppler effects is useful only in the case that the Doppler shift and width are large compared to the width $\Gamma\sim O(\lambda^2)\omega_L$; therefore it is natural to require $\beta\gg\lambda^2$. This condition supports the assumption that the resonance conditions (b) and (c) of Sec. II are not satisfied.

Now set $\Psi^{(0)}=\sum_{\alpha=s}^{f}C_\alpha(t)\phi_\alpha$ and write the two-level Schrödinger equation as

$$i\frac{\partial}{\partial t}C_\alpha=\sum_\beta H^{(2;2)}_{\alpha\beta}C_\beta, \tag{3.3}$$

where $\alpha,\beta=s,f$ and λ^2 has been reabsorbed into the matrix elements. The optical Bloch equations are obtained as follows: Set $C_s=e^{i\phi}B_s$, $C_f=e^{-i\phi}B_f$, and define

$$u=B_f^*B_s+B_fB_s^*,$$

$$v=-i(B_f^*B_s-B_fB_s^*),$$

$$w=|B_f|^2-|B_s|^2.$$

Then $\vec{r}=(u,v,w)$ satisfies the optical Bloch equations[22] in the "rotating frame":

$$d\vec{r}/dt=\vec{\Omega}\times\vec{r},\quad \vec{\Omega}=(2\Gamma S^2,0,-2\dot{\phi}+2\Delta_{fs}S^2), \tag{3.4}$$

where $\Delta_{fs}=\Delta_f-\Delta_s$ is the optical Stark shift of the resonance frequency.

One advantage of the Bloch equations is that any known solution of the one-photon problem can be used to construct solutions for the two-photon problem. For example the sech pulse[23] which corresponds to the effective field

$$\vec{\Omega}=\left(\frac{[1+(\nu\tau)^2]^{1/2}}{\tau}\operatorname{sech}\frac{t}{\tau},\ 0,\ \nu\tanh\frac{t}{\tau}\right)$$

leads to an exact analytical solution of (3.4). This solution can be used for the two-photon case by setting

$$2\Gamma=[1+(\nu\tau)^2]^{1/2}/\tau,$$

$$S(t)=[\operatorname{sech}(t/\tau)]^{1/2},$$

$$\dot{\phi}(t)=-\tfrac{1}{2}\nu\tanh(t/\tau)+\Delta_{fs}\operatorname{sech}(t/\tau).$$

With the initial conditions $u=v=0$ and $w=-1$, the solution for w is

$$w=\tanh(t/\tau),$$

which explicitly exhibits the complete population inversion of the two levels. Thus in the two-photon case what is needed is a $\operatorname{sech}^{1/2}$ pulse with an extra term in the frequency modulation to account for the time-dependent optical Stark shift.

Essentially complete inversion can also be achieved for other pulse shapes of greater experimental interest, such as a Gaussian pulse with linear chirping given by

$$S(t)=e^{-t^2/2\tau^2},\quad \dot{\phi}(t)=\mu t+\Delta_{fs}. \tag{3.5}$$

The constant term in $\dot{\phi}$ guarantees that the frequency has the correctly shifted value at pulse maximum. The effective field $\vec{\Omega}$ is

$$\vec{\Omega}=(2\Gamma e^{-t^2/\tau^2},\ 0,\ -2\mu t+2\Delta_{sf}-2\Delta_{sf}e^{-t^2/\tau^2}).$$

In the one-photon case complete inversion can be ensured by imposing the adiabaticity condition

$$\left|\frac{d\vec{\Omega}}{dt}\right|\ll|\vec{\Omega}|^2; \tag{3.6}$$

the vector $\vec{r}$ is then supposed to follow $\vec{\Omega}$ adiabatically. In the present problem (3.6) is not sufficient to ensure complete inversion since Ω_3 may change sign more than once during the pulse. Thus $\vec{r}$, which is adiabatically following $\vec{\Omega}$, would experience a decrease in the inversion w during part of the pulse. The various possibilities, which depend on the value of μ, are shown in Fig. 1. Inspection of the graph shows that there is a critical value μ_c such that for $\mu>\mu_c$, Ω_3 changes sign exactly once during the pulse. Thus if the adiabatic-following assumption (3.6) were satisfied for some $\mu>\mu_c$ (corresponding to line a in Fig. 1), complete inversion would be attained. On the other hand for $\mu<\mu_c$ complete inversion can fail even when adiabaticity is satisfied for most of the pulse. In this case Ω_3 changes sign three times (line b in Fig. 1). During the part of the pulse in which the first two sign changes occur, $\vec{r}$ can follow $\vec{\Omega}$ adiabatically. The inversion

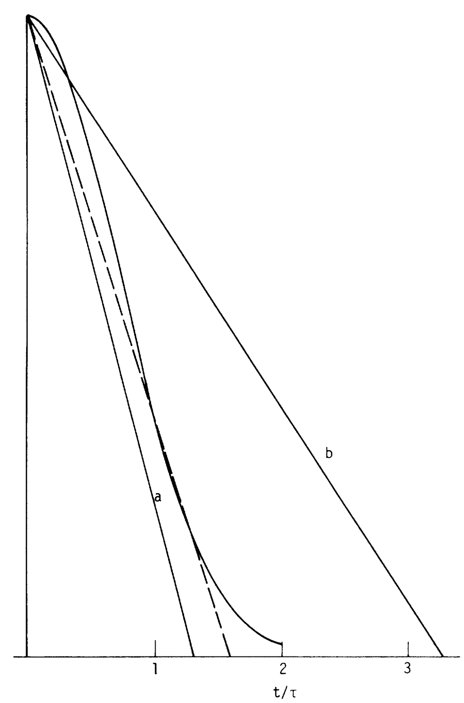

FIG. 1. Contributions to Ω_3 vs time in units of the pulse duration τ. The solid curve is $2\tau\Delta_{sf}e^{-t^2/\tau^2}$ and the straight lines are $(-2\mu t+2\Delta_{sf})\tau$ for different values of μ. In each case Ω_3 is the difference between the straight line and the curve. The dashed line represents the critical value of μ, while the solid lines a and b correspond respectively to supercritical and subcritical values.

increases between the first and second sign changes and decreases between the second and third. At the third sign change the field is weak so that $\vec{\Omega}$ is small and the adiabaticity condition (3.6) is violated. Thus $\vec{r}$ cannot follow $\vec{\Omega}$, and inversion cannot be restored.

IV. DOPPLER-FREE INVERSION IN Cs

A convenient example of two-photon resonance is the absorption of ruby laser light by atomic Cs. This experiment[24] has been performed (with an unchirped pulse) using the transition $6S_{1/2} \to 9D_{3/2}$ ($s \to f$ in the notation of Sec. III), with resonance energy $2\omega_L = 28\,860\ \mathrm{cm}^{-1}$. The intermediate states involved in (3.1) and (3.2) are restricted by dipole selection rules to $npP_{1/2}$, $npP_{3/2}$, and $nfF_{5/2}$; the sums were carried out to $n=14$. The necessary energies and dipole matrix elements were kindly provided by J. H. Scofield.[25] The shift and width are

$$\Gamma/\omega_L = 0.495\ \lambda^2 = 1.414\times10^{-14} I\,(\mathrm{W/cm^2}),$$

$$\Delta_{fs}/\omega_L = -2.038\ \lambda^2 = -5.823\times10^{-14} I\,(\mathrm{W/cm^2}),$$

where $\lambda = 1.7\times10^{-7}[I\,(\mathrm{W/cm^2})]^{1/2}$. The value of λ is obtained from the definition $\lambda = \mathcal{E}_0 \bar{d}/\omega_L$, where $\bar{d}$ is a representative value of the dipole matrix elements; in the present case $\bar{d} = 5D$.

Now consider the Gaussian, linearly chirped pulse (3.5) with strength $\lambda = 0.002$ ($I = 1.4\times10^8$ W/cm^2) and duration $\omega_L\tau = 2.5\times10^6$ ($\tau = 0.98$ nsec). The corresponding Rabi period is $\tau_R = \pi/\Gamma = 0.62$ nsec. These numbers have been chosen so that the intensity is below the dielectric breakdown value, and $\tau_R \cong \tau \ll T_1 \cong 35$ nsec, where T_1 is the estimated lifetime of the $D_{3/2}$ state under the experimental conditions quoted in Ref. 24.

The excited-state population resulting from a numerical solution of the Schrödinger equation (3.3) is shown in Fig. 2 for two values of $\mu\tau/\omega_L$: 2.5×10^{-6} and 6.25×10^{-6}. The former value is subcritical; it corresponds to line b in Fig. 1. The adiabaticity condition is well satisfied for most of the pulse ($|\dot{\vec{\Omega}}|/|\vec{\Omega}|^2 \cong 0.1$ at $t = 2.2\tau$), but inversion fails completely. The trouble is that Ω_3 has the wrong sign for most of the pulse; it regains the correct sign only after the field has become very small. The larger value of μ is supercritical, and essentially complete inversion is attained. Note that the oscillations in population for this case correspond to a large precession angle of $\vec{r}$ about $\vec{\Omega}$ so that small angle adiabatic following is definitely violated. The result for the critical value $\mu\tau/\omega_L = 5\times10^{-6}$ (not shown in Fig. 2) is a partial inversion with a final upper-level population of 0.6. For values of μ much larger than the critical value complete inversion fails again owing to the excessively nonadiabatic character of the pulses. It was found that complete inversion is possible for a range of μ values: $6\times10^{-6} \lesssim \mu\tau/\omega_L \lesssim 2\times10^{-5}$. The dimensionless parameter $\mu\tau/\omega_L$ is convenient because it is the fractional shift in frequency during the chirped pulse. Thus the important experimental question is the feasibility of generating chirped pulses in the range indicated above.

V. SUMMARY

The WGE theory was applied to nonmonochromatic radiation and specialized to excitation by two counterpropagating chirped pulses. It was shown that, in the absence of competing one-photon resonances, a multilevel system is reduced (in first approximation) to an equivalent two-level system. Qualitative differences between one- and two-photon pulse excitations are that for the latter (1) Doppler broadening can be eliminated and (2) population inversion can fail even with adiabatic following. Population inversion was explicitly computed for the $6S_{1/2} \to 9D_{3/2}$ two-photon transition in Cs. Almost complete inversion resulted for chirp rates satisfying $6\times10^{-6} \lesssim \mu\tau/\omega_L \lesssim 2\times10^{-5}$.

ACKNOWLEDGMENTS

We thank J. H. Scofield for kindly supplying computed energy levels and transition matrix of atomic Cs for the computations of Sec. IV. The numerical differential equation solver used in Sec. IV is a version of a computer program written by C. W. Gear and modified by Alan Hindmarsh.

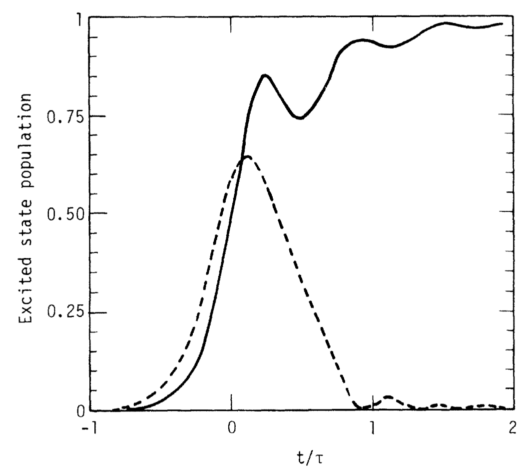

FIG. 2. Excited-state population vs time, in units of pulse duration τ. The intensity and pulse duration are $I = 1.4\times10^8$ W/cm^2 and $\tau = 0.98$ nsec, respectively. The solid curve corresponds to $\mu\tau/\omega_L = 6.25\times10^{-6}$ (line a in Fig. 1) and the dashed curve to $\mu\tau/\omega_L = 2.5\times10^{-6}$ (line b in Fig. 1).

*Work performed under the auspices of the U. S. Energy Research and Development Administration.

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[21]In this language the calculations presented in Refs. 19 and 20 correspond to a nonvanishing $H^{(1;1)}$ which is used to determine the t_1 dependence; the t_2 dependence is neglected.

[22]Equations (3.4) are equivalent to Eq. (42) of Ref. 2; where the vector $\vec{\Omega}$ defined in (3.4) corresponds to their $\vec{\gamma}$. In that paper the Bloch equations are obtained as the result of two successive unitary transformations. The first leads to a time-dependent basis set in which the transformed Hamiltonian has matrix elements joining ϕ_s to ϕ_f (ϕ_2 to ϕ_1 in Ref. 2). This is equivalent to the calculation of $H^{(2;2)}$ in Sec. II. The second transformation is the familiar one to the "rotating frame" given here by the change $C_\alpha \rightarrow B_\alpha$ defined just below (3.3).

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