Phase-dependent pump-probe line-shape formulas

J. H. Eberly and V. D. Popov*
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
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Pump-probe interactions underlie the physics of four-wave and other wave-mixing effects. We present analytic formulas for probe response in a special category of pump-probe experiments. We consider a two-level system excited by two distinct fields, the stronger field with fixed frequency and the weaker with a tunable frequency. We focus attention on the spectral regions near the Rabi sidebands of the strong field and its various subharmonics. We consider times sufficiently short that relaxation effects can be ignored, but sufficiently long to encompass many cycles of population oscillation between the ground and excited states. Because there is no damping, both fields are in this sense strong, and we report here the time average of the response. Our results are obtained by making a suitable second-order rotating wave approximation and are shown to involve the initial phase relation between the two fields as well as the initial angles of orientation of the two-level system's Bloch vector. Depending on these initial phase and orientation angles, the absorption line shape may be either positive or negative and either symmetric or antisymmetric.

I. INTRODUCTION

The many applications of wave mixing in nonlinear and quantum optics make the underlying two-field interaction of widespread interest. In a more general sense all nonlinear wave-mixing interactions illustrate the richness and complexity of pump-probe physics. We study a particular set of pump-probe phenomena in this paper. We consider the situation shown in Fig. 1, in which a two-level quantum system is irradiated by two radiation fields simultaneously, but generally not at the same frequency. To facilitate the writing we will refer to the system as an atom and the two fields as if they came from two separate lasers. Other similar situations to which the model is applicable are obvious and need not be detailed.

Much work has been devoted in the recent quantum optical literature to two- and three-level atoms excited in a pump-probe configuration, and we cannot hope to cite all significant papers. A few that we have encountered or that have been recommended to us are listed in Ref. 1. Our attention here will be directed to a situation not covered in these citations, but a situation that is probably accessible to experimental study. We refer to the case in which observations are made in a time rather short compared with relevant relaxation times but still long compared with the periods of various population oscillations.

Our specific domain of interest is easily described by analogy with a one-laser line-shape formula. Consider Fig. 1 again but remove the probe laser. The probability of occupying the excited state of the atom, in the absence of relaxation, is well known to be given by

$$P_2(t)=\frac{r^2}{\Delta^2+r^2}\sin^2(\Omega/2)t \ . \tag{1.1}$$

Here Δ is the detuning of the strong laser, r is our symbol for the Rabi frequency on resonance, and Ω is the detuned Rabi frequency

$$\Delta=\omega_{21}-\omega_L, \quad r=(2dE_L/\hbar), \quad \Omega=[(r^2+\Delta^2)]^{1/2} \ , \tag{1.2}$$

where we have taken the laser field to be

$$E=E_L e^{-i\omega_L t}+\text{c.c.} \tag{1.3}$$

and d is the transition's dipole matrix element.

Over many cycles of Rabi oscillation the steady-state average excitation probability is clearly given by

$$\langle P_2\rangle=\frac{r^2/2}{\Delta^2+r^2} \ , \tag{1.4}$$

and one can say that (1.4) exhibits the resonant response of the atom to the field and that the linewidth is due solely to power broadening. This is, of course, not the same as the steady-state response formula, which involves the atoms' relevant relaxation parameters in the well-known way:

$$P_2(\infty)=\frac{r^2T_1/2T_2}{\Delta^2+(1/T_2)^2+r^2T_1/T_2} \ . \tag{1.5}$$

In this paper we will obtain the formulas corresponding to (1.4) when the probe laser shown in Fig. 1 is included. The relevant line-shape variable will be the de-

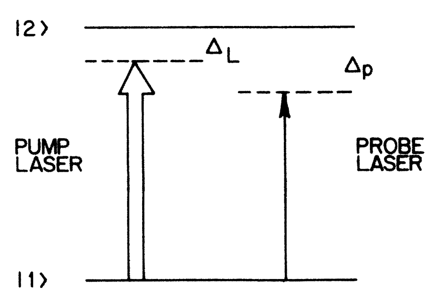

FIG. 1. Two-level quantum system irradiated by two near-resonant fields simultaneously.

tuning of the probe laser, not the strong pump laser. Although these formulas are not difficult to obtain, they do not seem to appear in the literature.

II. MATHEMATICAL FORMULATION

The absence of relaxation means that density matrix methods are unnecessary, and we will concentrate on the time dependence of the state vector amplitudes associated with the upper and lower levels of the atom. The Hamiltonian can be rewritten several ways:

$$H = H_0 - dE_{\text{probe}} \,, \tag{2.1a}$$

$$H = H_A - dE_L - dE_p \,, \tag{2.1b}$$

where H_A is the usual bare atomic Hamiltonian and H_0 is the "large" part of the total Hamiltonian, containing both the bare atom and the interaction dE_L with the pump laser.

We adopt the usual rotating wave approximation (RWA) and drop the "counter-rotating" parts of the Hamiltonian at the outset, in which case the Hamiltonian can be written

$$(H_0+H_p)_{\text{RWA}} = \begin{bmatrix} E_{21}/2 & -(r/2)e^{-i\omega_L t - i\psi_L} - (r_p/2)e^{-i\omega_p t - i\psi_p} \\ -(r/2)e^{i\omega_L t + i\psi_L} - (r_p/2)e^{i\omega_p t + i\psi_p} & -E_{21}/2 \end{bmatrix} . \tag{2.2}$$

Here E_{21} is the bare atomic transition energy, and we have located the zero of energy half way between the two bare energy levels. We have indicated explicitly the phases[2] of the two laser fields at $t=0$, and we have dropped from r_L the subscript L for convenience. In the usual rotating frame, determined by the pump-laser frequency, the two-level atom's Schrödinger equation can now be written

$$i\begin{bmatrix} \dot c_2 \\ \dot c_1 \end{bmatrix} = \begin{bmatrix} \dfrac{\Delta}{2} & -\dfrac{r}{2} \\ -\dfrac{r}{2} & -\dfrac{\Delta}{2} \end{bmatrix}\begin{bmatrix} c_2 \\ c_1 \end{bmatrix} + \begin{bmatrix} 0 & -\dfrac{r_p}{2}e^{i(\Delta_p t+\psi)} \\ -\dfrac{r_p}{2}e^{-i(\Delta_p t+\psi)} & 0 \end{bmatrix}\begin{bmatrix} c_2 \\ c_1 \end{bmatrix} , \tag{2.3}$$

where we have defined Δ_p to be the frequency offset of the probe from the pump, and ψ to be the relative phase

$$\Delta_p = \omega_L - \omega_p, \quad \psi = \psi_L - \psi_p \,. \tag{2.4}$$

The large part of the Hamiltonian is now time independent,

$$H_0 = \begin{bmatrix} \Delta/2 & -(r/2) \\ -r/2 & -\Delta/2 \end{bmatrix} , \tag{2.5}$$

and its eigenstates provide a convenient basis, as could be expected. These are designated by $|+\rangle$ and $|-\rangle$, and their corresponding amplitudes by $a_+(t)$ and $a_-(t)$, which are related to $c_2(t)$ and $c_1(t)$ by a rotation matrix $R(\theta)$:

$$\begin{bmatrix} c_2 \\ c_1 \end{bmatrix} = R(\theta)\begin{bmatrix} a_+ \\ a_- \end{bmatrix} = \begin{bmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{bmatrix}\begin{bmatrix} a_+ \\ a_- \end{bmatrix} , \tag{2.6}$$

where the angle θ is defined in a standard way:

$$r/\Omega = \sin 2\theta, \quad \Delta/\Omega = \cos 2\theta \,, \tag{2.7}$$

and the detuned Rabi frequency Ω has already been defined in (1.2).

In the dressed basis the probe part of the total Hamiltonian has four nonzero matrix elements, and the equations obeyed by the a's are

$$i\dot a_+(t) = (\Omega/2)a_+(t) + V_{++}(t)a_+(t) + V_{+-}(t)a_-(t) \,,$$
$$i\dot a_-(t) = -(\Omega/2)a_-(t) + V_{-+}(t)a_+(t) + V_{--}(t)a_-(t) \,, \tag{2.8}$$

where the V's are given by (with $\beta = \Delta_p t + \psi$)

$$V_{++} = (r_p/2)\sin 2\theta \cos\beta \,, \tag{2.9a}$$

$$V_{+-} = -(r_p/2)(\cos 2\theta \cos\beta + i\sin\beta) \,, \tag{2.9b}$$

$$V_{-+} = (V_{+-})^* \,, \tag{2.9c}$$

$$V_{--} = -V_{++} \,. \tag{2.9d}$$

The explicit equations for the a's are therefore given by

$$i\dot a_+ = \left[\frac{\Omega}{2} + \frac{r_p}{2}\sin 2\theta\cos\beta\right]a_+ - \frac{r_p}{2}(\cos 2\theta\cos\beta + i\sin\beta)a_- \,, \tag{2.10a}$$

$$i\dot a_- = \left[-\frac{\Omega}{2} - \frac{r_p}{2}\sin 2\theta\cos\beta\right]a_- - \frac{r_p}{2}(\cos 2\theta\cos\beta - i\sin\beta)a_+ \,. \tag{2.10b}$$

Well-known methods are available for dealing with such equations.[3] In Sec. III we will restrict our attention to the most important case, in which the pump field frequency is fixed at the bare atomic transition frequency. In this case $\Delta=0$ and so $\cos 2\theta = 0$, thus simplifying Eqs. (2.10) slightly.

III. SOLUTION OF AMPLITUDE EQUATIONS FOR RESONANT PUMP

When the pump is resonant, Eqs. (2.10) take the form

$$i\dot{a}_+ = \left[\frac{r}{2} + \frac{r_p}{2}\cos(\Delta t+\psi)\right]a_+ - i\frac{r_p}{2}\sin(\Delta t+\psi)a_- , \quad (3.1a)$$

$$i\dot{a}_- = \left[-\frac{r}{2} - \frac{r_p}{2}\cos(\Delta t+\psi)\right]a_- + i\frac{r_p}{2}\sin(\Delta t+\psi)a_+ . \quad (3.1b)$$

The diagonal time dependences in Eqs. (3.1) can be integrated. This is the same as shifting the phases of the amplitudes so we introduce new variables b_+ and b_- as follows:

$$b_\pm(t) = a_\pm(t)e^{\pm i(r_p/2\Delta)\sin(\Delta t+\psi)} . \quad (3.2)$$

We have further simplified the notation now that there is no pump detuning by using Δ without a suffix to mean the probe detuning. It will be the interesting spectroscopic parameter in this paper. The equations for the b's are

$$i\dot{b}_+(t) = (r/2)b_+(t) - i\sin(\Delta t+\psi)(r_p/2)b_-(t)e^{i(r_p/\Delta)\sin(\Delta t+\psi)} , \quad (3.3a)$$

$$i\dot{b}_-(t) = -(r/2)b_-(t) + i\sin(\Delta t+\psi)(r_p/2)b_+e^{-i(r_p/\Delta)\sin(\Delta t+\psi)} . \quad (3.3b)$$

It is now useful to introduce the Bessel function expansion

$$e^{i(r_p/\Delta)\sin(\Delta t+\psi)} = \sum_{n=-\infty}^{+\infty} e^{in(\Delta t+\psi)}J_n(r_p/\Delta) \quad (3.4)$$

and write the sine functions as exponentials as well. Then the equations become

$$i\dot{b}_+(t) = \frac{r}{2}b_+(t) - \frac{r_p}{4}(e^{i(\Delta t+\psi)} - e^{-i(\Delta t+\psi)}) \times \sum_{n=-\infty}^{+\infty} e^{in(\Delta t+\psi)}J_n(r_p/\Delta)b_-(t) , \quad (3.5a)$$

$$i\dot{b}_-(t) = -\frac{r}{2}b_-(t) + \frac{r_p}{4}(e^{i(\Delta t+\psi)} - e^{-i(\Delta t+\psi)}) \times \sum_{n=\infty}^{\infty} (-1)^n e^{in(\Delta t+\psi)}J_n(r_p/\Delta)b_+(t) . \quad (3.5b)$$

Equations (3.5) imply the existence of an infinite sequence of resonances at well-defined probe frequencies associated with the pump's Rabi sidebands and their subharmonics. We will concentrate here on the nature of the phase dependence of the pump-probe process and deal explicitly only with the principal sideband resonance. In this case, as we show below, it is appropriate to assume that only J_0 makes an appreciable contribution to the sums because $r_p/\Delta \ll 1$. Of course $J_0=1$ in the limit of small argument, so no evidence of the Bessel functions remains under our (much more severe than necessary) assumption. The b equations reduce to

$$i\dot{b}_+(t) = \frac{r}{2}b_+(t) + \frac{r_p}{4}e^{-i(\Delta t+\psi)}b_-(t) , \quad (3.6a)$$

$$i\dot{b}_-(t) = -\frac{r}{2}b_-(t) + \frac{r_p}{4}e^{i(\Delta t+\psi)}b_+(t) . \quad (3.6b)$$

In this form the b equations are exactly like the starting equations for the c's of Sec. II. In effect we have made a second rotating wave approximation, and the remaining physics is not just slowly varying but *very* slowly varying whenever Δ is approximately equal to r, i.e., whenever the probe frequency is in the neighborhood of the principal Rabi sideband.

We now write the equations appropriate to the second rotating frame, which is evidently moving at the frequency Δ. We introduce the very slowly varying amplitudes B_+ and B_- by the definitions

$$b_\pm = B_\pm e^{\pm i(\Delta t+\psi)/2} . \quad (3.7)$$

We also introduce the abbreviations

$$\mu = \tfrac{1}{2}(r-\Delta), \quad \kappa = \frac{r_p}{4} \quad (3.8)$$

and find the equations[4] (see Fig. 2)

$$i\dot{B}_+(t) = \mu B_+ + \kappa B_- , \quad (3.9a)$$

$$i\dot{B}_-(t) = -\mu B_- + \kappa B_+ . \quad (3.9b)$$

What we have really done in proceeding from Eqs. (3.5) to Eqs. (3.9) has been to isolate the harmonic components of b_+ and b_- that have very slow amplitudes in the neighborhood of $\Delta = r_p$ and ignore the other components because they are relatively much faster and so average quickly to zero. For other detuning values, notably at the Rabi subharmonics $\Delta = r_p/n$, other harmonic components of the b's are more important. In any event, we can now see that in the neighborhood of $\Delta = r_p$ our assumptions are justified.

Equations (3.9) are easily solved. As in Sec. II the equations can be diagonalized by a simple rotation:

$$\begin{bmatrix} B_+(t) \\ B_-(t) \end{bmatrix} = R(\alpha)\begin{bmatrix} e^{-i\bar{\mu}t} & 0 \\ 0 & e^{i\bar{\mu}t} \end{bmatrix} R^{-1}(\alpha)\begin{bmatrix} B_+(0) \\ B_-(0) \end{bmatrix} , \quad (3.10)$$

where $\bar{\mu}$ and $-\bar{\mu}$ are the eigenvalues of the second RWA Hamiltonian

$$\bar{\mu} = (\mu^2 + \kappa^2)^{1/2} . \quad (3.11)$$

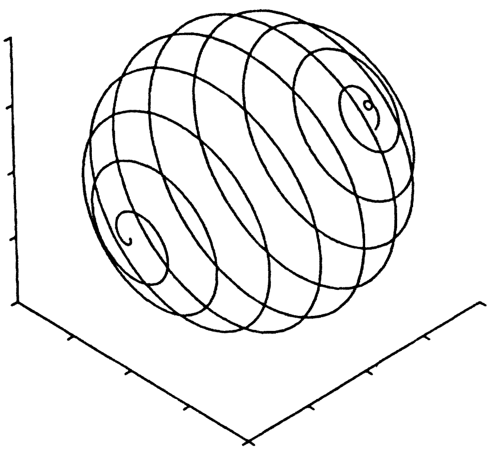

FIG. 2. Trajectory of the Bloch vector in the usual rotating frame (rotating about the vertical axis of the figure at the frequency of the pump laser). In this frame the motion is still almost entirely a rotation, although about a perpendicular axis, so it is clear that a second rotating transformation is appropriate.

The appropriate rotation angle α and rotation matrix $R(\alpha)$ are now given by

$$\mu/\bar{\mu}=\cos 2\alpha, \quad \kappa/\bar{\mu}=\sin 2\alpha \ , \tag{3.12}$$

$$R(\alpha)=\begin{bmatrix} \cos\alpha & \sin\alpha \\ -\sin\alpha & \cos\alpha \end{bmatrix} . \tag{3.13}$$

IV. AVERAGE INVERSION

The solutions found for the B's allow the solutions for the original c's to be found via (2.6), (3.2), and (3.7). We will regard either the inversion or the upper-state population as observable, and so compute $|c_2(t)|^2$. We easily find first

$$c_2(t)=\cos\theta B_+(t)e^{-i\Phi(t)}+\sin\theta B_-(t)e^{i\Phi(t)} \ , \tag{4.1}$$

where the B's are obtained from (3.10) and $\Phi(t)$ is the phase factor obtained from (3.2) and (3.7) together,

$$\Phi(t)=\beta+(r_p/\Delta)\sin\beta \ , \tag{4.2}$$

where $\beta=\Delta t+\psi$, as before. Obviously we can obtain

$$|c_2(t)|^2=\cos^2\theta\,|B_+|^2+\sin^2\theta\,|B_-|^2 + 2\sin\theta\cos\theta\,\mathrm{Re}(B_+^*B_-e^{2i\Phi}) \ . \tag{4.3}$$

Some simplifications can be effected immediately. We will continue to assume that the probe is tuned near the pump's Rabi sideband, so that the B's are very slow variables. Thus we can discard all of the harmonic frequencies $n\Delta$ for $n\neq 0$ in the factor $e^{2i\Phi}$ since they are much more rapidly varying. Under the same pump resonance condition adopted earlier we have $\theta=\pi/4$. Thus (4.3) reduces to

$$|c_2|^2=\tfrac{1}{2}+(r_p/\Delta)\mathrm{Re}(B_+^*B_-) \ , \tag{4.4}$$

where the factor r_p/Δ comes from the leading contribution of the harmonic amplitude $J_1(r_p/\Delta)$.

If desired, both the lower state population $|c_1|^2$ and the inversion $|c_2|^2-|c_1|^2$ are easily computed from (4.4) by using probability conservation $|c_1|^2+|c_2|^2=1$. The inversion w is perhaps more interesting since it is also one of the components of the two-level atom's Bloch vector. We record the result

$$w=|c_2|^2-|c_1|^2=2(r_p/\Delta)\mathrm{Re}(B_+^*B_-) \ . \tag{4.5}$$

Now we compute the time average of this expression. We easily find from (3.10) the result

$$w=(r_p/\Delta)\sin 2\alpha[\cos 2\alpha(|B_{+0}|^2-|B_{-0}|^2) + \sin 2\alpha(2\,\mathrm{Re}B_{+0}^*B_{-0})] \ . \tag{4.6}$$

The initial values of the B's have to be related to the initial values of the c's. This is easily done through (2.6), (3.2), and (3.7). One finds

$$B_{+0}^*=(\eta^*\cos\theta-\xi^*\sin\theta)e^{-i\Phi_0} \ , \tag{4.7a}$$

$$B_{-0}=(\eta\sin\theta+\xi\cos\theta)e^{-i\Phi_0} \ , \tag{4.7b}$$

where we have defined

$$\xi=c_1(0), \quad \eta=c_2(0) \ . \tag{4.8}$$

Again we consider only pump resonance ($\theta=\pi/4$) and so find

$$2\,\mathrm{Re}(B_{+0}^*B_{-0})=v_0\sin 2\Phi_0+w_0\cos 2\Phi_0 \ , \tag{4.9a}$$

$$|B_{+0}|^2-|B_{-0}|^2=-u_0 \ , \tag{4.9b}$$

where we have introduced the usual notation for the components of the initial Bloch vector

$$u_0=2\,\mathrm{Re}(\xi^*\eta) \ , \tag{4.10a}$$

$$v_0=2\,\mathrm{Im}(\xi^*\eta) \ , \tag{4.10b}$$

$$w_0=|\eta|^2-|\xi|^2 \ . \tag{4.10c}$$

Thus we finally have a compact expression for the time-averaged inversion,

$$w=(r_p/\Delta)\sin 2\alpha[u_0\cos 2\alpha+(-v_0\sin 2\psi + w_0\cos 2\psi)\sin 2\alpha] \ . \tag{4.11}$$

We have used (4.2) at $t=0$ for Φ_0 and have retained only the lowest-order contribution $\Phi_0=\psi$, consistent with our earlier expansion of $J_1(r_p/\Delta)$.

The final result shows that the time-average inversion is very small, which was to be expected since the strong resonant pump tends to distribute population equally between the two levels. In addition, we see that the inversion can be written as a vector projection. We define a unit vector $\mathbf{A}$ as follows:

$$A_1=\cos 2\alpha \ , \tag{4.12a}$$

$$A_2=-\sin 2\psi\sin 2\alpha \ , \tag{4.12b}$$

$$A_3=\cos 2\psi\sin 2\alpha \ . \tag{4.12c}$$

Then (4.11) can be written

$$w=(r_p/\Delta)\sin 2\alpha\mathbf{S}_0\cdot\mathbf{A} \ , \tag{4.13}$$

where $\mathbf{S}_0$ is the initial Bloch vector $\mathbf{S}_0=(u_0,v_0,w_0)$.

V. LINE SHAPE

Expressions (4.11) and (4.13) depend on probe detuning, and either one can serve as our primary line-shape formula. To see the dependence of (4.11) on probe detuning we must recall (3.8) and (3.12). Then it is obvious that (4.11) can be rewritten in the form

$$w(x)=(r_p/\Delta)\left[\frac{K+Lx}{x^2+1}\right], \tag{5.1}$$

where x, the new detuning variable, is centered on the first sideband resonance:

$$x=\mu/\kappa=(\Delta-r)/(r_p/2) \tag{5.2}$$

and the coefficients are

$$K=-v_0\sin 2\psi+w_0\cos 2\psi , \tag{5.3a}$$

$$L=u_0 . \tag{5.3b}$$

To remain consistent with earlier assumptions we have to restrict application of (5.1) to values of Δ in the neighborhood of r. This is just what is wanted, however, since now it is shown explicitly that $\Delta=r$ is the center of a narrow resonance line.

The shape of the line is exactly Lorentzian, with halfwidth equal to $r_p/2$, unless L is nonzero. In this case there is a component of dispersion shape mixed into the "normal" absorption profile. It is even possible for the line shape to be completely "dispersive" if $K=0$. This occurs when $v_0=w_0=0$, in other words when $|u_0|=1$.

We can identify the two different pure dispersive or absorptive line shapes easily in terms of the initial states. For example, $K=0$ corresponds to an initial condition in which the atom is prepared in one of its dressed states at the outset, precisely the dressed state that cannot be reached by resonant pump excitation. It leads to a pure dispersive line shape. Conversely, if the initial state is prepared by resonant pumping from the ground state, then only the v and w components of the Bloch vector can be involved and necessarily $L=u_0=0$. Then the shape is purely absorptive.

The sign of the inversion is also predicted reasonably. Suppose that the initial state is prepared in the "most natural" way, i.e., by allowing the atom to relax over many lifetimes to its ground state. In this case $w_0=-1$ and $u_0=v_0=0$. If $\psi=0$, we predict $w<0$, indicating that the probe is absorbed, as expected. However, if coherent excitation from the ground state with a resonant pump laser prepares $w_0=+1$, then we predict $w>1$, indicating that the prepumped atom is able to amplify the probe.

The role of a nonzero value of the relative pump-probe phase is less obvious. Consider the case where the atom begins in its ground state, but $\psi=\pi/4$. In this case we predict $w=0$, completely independent of the probe tuning. In other words *there are cases with no line shape at all.* Recall that this does not mean that there is no interaction; there is simply no time-independent interaction.

VI. SUMMARY

We have solved the Schrödinger equation for a two-level atom driven by pump and probe fields simultaneously. Our solution (3.10) is nonperturbative, and thus allows the derivation of the time-average loss-free line-shape formula (5.1). Our result is not exact, of course, but depends on the validity of a second rotating frame transformation. There are infinitely many ways to choose the second frame, and we have dealt explicitly only with the frame identified with the "primary" pump-probe resonance which occurs when the probe is tuned away from the atomic resonance by one unit of pump Rabi frequency.

We have shown that the probe line shape (5.1) depends on the initial phase difference between the pump and probe lasers and on the orientation of the initial Bloch vector. Depending on these parameters, the line shape can be absorptive or dispersive or can be absent altogether. We have interpreted some of these results in a physical way.

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*Permanent address: Department of General Physics, Engineering Physics Institute, Kashirskoe Shosse, 31, Moscow 115409, U.S.S.R.

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[3]The classic references are S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955) and J. H. Shirley, *ibid.* **138**, B979 (1965). See also, for example, Sec. 2.3 of S. Stenholm, *Foundations of Laser Spectroscopy* (Wiley, New York, 1984).

[4]The B amplitude equations are equivalent to Bloch equations in a doubly rotating frame. The behavior of the Bloch vector under the influence of both pump and probe beams, computed numerically in the usual rotating frame, is shown in Fig. 2. We will discuss pump-probe dynamics of two-level systems in more detail elsewhere, using the Bloch vector approach.