## Propagation of modulated optical fields through saturable-absorbing media: a general theory of modulation spectroscopy

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Received February 27, 1985; accepted May 30, 1985

The propagation of a weakly modulated light beam through a nonlinear material is treated. The optical field representing such a beam consists of a strong carrier-frequency component and two weak, symmetrically displaced sidebands that combine to form a field, which may be purely amplitude modulated (AM), frequency modulated (FM), or some combination of these two modulation forms. It is shown that, for any optical nonlinearity, two modulation forms exist that have the property that the form of modulation is invariant under propagation of the beam. If a modulation form other than one of these natural modes is injected into the nonlinear medium, the modulation form will change as the beam propagates, asymptotically approaching the natural mode that experiences the lower attenuation. Explicit expressions for these natural modes are presented for the case in which the nonlinear medium can be modeled as a collection of two-level atoms. For the special case of an on-resonance pump beam, the natural modes correspond to pure amplitude modulation and pure frequency modulation. This formalism provides a general description of saturation spectroscopy for both AM and FM fields. Formulas are derived for the rate at which a pure FM beam is converted to an AM beam owing to its interaction with a two-level atomic medium. We also consider the variation that is due to propagation of the depth of modulation of an AM wave interacting resonantly with two-level atoms. The formalism predicts the existence of spectral features whose shape depends sensitively on the internal relaxation processes of the material. Experimental spectra are presented for ruby, alexandrite, and fluorescein in glass and are interpreted.

#### 1. INTRODUCTION

A standard technique in nonlinear optical spectroscopy is to measure the modification of the transmission of a weak probe beam through a sample that is saturated by a strong pump beam.<sup>1-5</sup> The results of such measurements have been used to obtain an understanding of the electronic and structural properties of materials and to probe the dynamics of relaxation processes.<sup>6</sup> These measurements are often made with pump and probe beams derived from separate laser sources. An alternative procedure is to use only a single laser and to create the probe beam by passing the monochromatic laser beam through an optical modulator.<sup>7–15</sup> The unmodulated portion of the laser beam then acts as the pump beam, while the modulation sidebands form the probe.<sup>16–19</sup> When such a modulated beam interacts with a nonlinear material, the form of modulation changes. The detection and determination of this change are the basis of modulation spectroscopy. An advantage of modulation spectroscopy over the use of separate laser sources is that at most one stabilized laser is required. In fact, under certain circumstances, the results of such an experiment depend much more strongly on the modulation frequency than on the laser frequency, and under these conditions the frequency stability of the laser is of only minor importance.

In this paper we present an analysis of the propagation of a weakly modulated beam through a saturable-absorbing medium. It provides a unified treatment of the various forms of modulation spectroscopy. We begin by showing in Section 2 that such a beam can always be represented as the superposition of a pump component at frequency  $\omega_0$  and two weak components at frequencies  $\omega_0 \pm \delta \omega$ , where  $\delta \omega$  represents the modulation frequency. One special case involves a monochromatic (or single-sideband) probe beam of frequency  $\omega_0$ +  $\delta\omega$ . Because of the nonlinearity of the material response, this sideband is strongly coupled to the symmetrically displaced sideband at frequency  $\omega_0 - \delta \omega$ .<sup>20-22</sup> As the beam propagates, the amplitude of this sideband grows from its initially zero value, and the beam develops some complex form of modulation. Similarly, an arbitrarily modulated input beam changes its type of modulation as it propagates, since the two single-sideband components will in general experience different attenuations and dispersive phase shifts. However, we show in Section 3 that for any arbitrary nonlinear medium, for any pump beam, and for any modulation frequency, there always exist two modulation forms that are left invariant as the beam propagates. We refer to these modulation forms as the natural modes of propagation of the modulated optical field. These natural modes can be usefully represented by either of two basis sets. In Section 3 we represent these modes as a linear combination of single-sideband (i.e., monochromatic) fields, whereas in Section 4 we represent them as a linear combination of amplitude-modulated (AM) and frequency-modulated (FM) fields. The treatment of Section 3 deals with an arbitrary nonlinear optical medium and demonstrates that natural modes of modulation exist for any such medium. Section 4 illustrates the use of the AM-FM basis set for the important special case of a medium that can be modeled as a collection of two level atoms.<sup>23</sup> For such a medium, it is particularly useful to represent the natural modes in the AM-FM basis set, since the Bloch vector formalism for calculating the atomic response naturally treats the AM and FM components separately. In Section 5 we consider representative examples of the natural-mode solutions. The application of this natural-mode formalism to saturation spectroscopy is presented in Sections 6 and 7. In Section 6 we calculate the rate at which the FM component is converted to an AM component as the beam propagates. This conversion provides the basis of FM saturation spectroscopy. In Section 7 we consider a spectroscopic technique that we call AM saturation spectroscopy in which the change in the depth of modulation of an AM beam is measured. Since only the AM component of the light beam is of relevance, this technique is largely insensitive to the presence of FM noise from the laser. This technique provides a sensitive probe for measuring the population relaxation processes of a saturable medium

#### 2. DEFINITION OF THE MODULATED FIELD

The theory developed in this paper makes use of several different electric-field decompositions, each of which is appropriate for describing some particular aspect of the propagation of modulated optical fields. The most common decomposition in nonlinear optics is in terms of the Fourier amplitudes of the field. Using this notation, a weakly modulated optical field E(z, t) can be represented as the sum of three monochromatic field components by

 $E(z,t) = E_0(z,t) + E_{+\delta\omega}(z,t) + E_{-\delta\omega}(z,t),$ 

$$E_0(z, t) = F_0(z)\exp(i\omega_0 t) + \text{c.c}$$
(2.2a)

$$= A_0(z) \exp[i(\omega_0 t - k_0 z)] + \text{c.c.}, \quad (2.2b)$$

with  $k_0 = \omega_0/c$ , and where

$$E_{\pm\delta\omega}(z,t) = F_{\pm\delta\omega}(z)\exp[i(\omega_0 \pm \delta\omega)t] + c.c \qquad (2.3a)$$

 $= A_{\pm\delta\omega}(z)\exp\{i[(\omega_0\pm\delta\omega)t - (k_0\pm\delta k)z]\} + \text{c.c.}, \quad (2.3b)$ 

with  $\delta k = \delta \omega/c$ . Here the quantities  $F_0(z)$  and  $F_{\pm\delta\omega}(z)$  are the temporal Fourier amplitudes of the field, which show a rapid spatial variation, whereas the quantities  $A_0(z)$  and  $A_{\pm\delta\omega}(z)$  are the slowly varying amplitudes from which this rapid spatial variation has been factored out. The quantities  $E_{\pm\delta\omega}$  represent the weak sidebands, symmetrically detuned by frequency  $\pm\delta\omega$  ( $\delta\omega \ll \omega_0$ ) from the strong pump wave  $E_0$  at frequency  $\omega_0$ . These sidebands are assumed weak in the sense that the material response is a linear function of  $F_{\pm\delta\omega}$ . This decomposition of the field into its frequency components is most usefully employed when the pump-sideband detuning  $\delta\omega$  is sufficiently large that conventional spectrometers can be used to separate the total field into its spectral components.

Conversely, when the pump-sideband detuning is small (typically < 1 GHz), it is often possible to measure directly the beat frequency between the various spectral components of the total field. In this case it is convenient to express the electric field in a manner that displays the resulting temporal modulation of the total field:

$$E(z,t) = 2 \operatorname{Re}(\tilde{\mathcal{E}}(z,t) \exp\{i[\omega_0 t - k_0 z + \phi_0(z)]\}), \quad (2.4)$$

where

$$\tilde{\mathscr{E}}(z,t) = \mathscr{E}_0(z) + \delta \tilde{\mathscr{E}}(z,t). \tag{2.5}$$

(Wherever the possibility of ambiguity exists, tildes will be

used to denote complex quantities.) Here  $\delta \tilde{\mathcal{E}}(z, t)$  and  $\mathcal{E}_0(z)$  represent the weak modulated component and the unmodulated (i.e., carrier-wave) component of the total field, respectively.  $\phi_0(z)$  represents the phase of the carrier wave and is selected so that  $\mathcal{E}_0(z)$  is everywhere real. It is convenient to decompose the weak field  $\delta \tilde{\mathcal{E}}(z, t)$  into its real and imaginary parts as

$$\delta \mathcal{E}(z,t) = \delta \mathcal{E}_{AM}(z,t) + i \delta \mathcal{E}_{FM}(z,t). \qquad (2.6)$$

This notation has been adopted because, as shown in Fig. 1, the real and the imaginary parts of  $\delta \tilde{\mathcal{E}}(z, t)$  give rise, respectively, to amplitude and to phase modulation of the total field. In fact, the intensity associated with the field given by Eqs. (2.4)–(2.6) is given by

$$I(z) = \frac{c}{4\pi} \langle E(z,t)^2 \rangle_t = \frac{c}{2\pi} |\tilde{\mathscr{E}}(z,t)|^2 = \frac{c}{2\pi} [\mathscr{E}_0(z)^2 + 2\mathscr{E}_0(z)\delta\mathscr{E}_{\rm AM}(z,t)].$$
(2.7)

Hence only the real part of  $\delta \tilde{\mathcal{E}}(z, t)$  contributes to the temporal modulation of the total intensity.

For the case under consideration in which the modulation is due to two symmetrically displaced sidebands [Eq. (2.1)], the probe field can be represented as

$$\delta \hat{\mathcal{E}}(z,t) = \delta \mathcal{E}(z) (\cos \psi \exp\{i[\delta \omega t - \delta k z + \phi_1(z)]\} + \sin \psi \exp\{-i[\delta \omega t - \delta k z + \phi_1(z) + \theta]\}, \quad (2.8a)$$

or equivalently as

(2.1)

$$\delta \tilde{\mathscr{E}}(z,t) = \delta \mathscr{E}(z) \exp(-i\theta/2) \times (\cos \psi \exp\{i[\delta \omega t - \delta k z + \phi_1(z) + \theta/2]\} + \sin \psi \exp\{-i[\delta \omega - \delta k z + \phi_1(z) + \theta/2]\}),$$
(2.8b)

where  $\phi_1(z)$  denotes the spatial variation of the phase of the sidebands that is due to dispersion. Equations (2.4), (2.5), and (2.8) represent an arbitrary form of modulation of the total electric field. The actual form of modulation is specified by the parameters  $\psi$  and  $\theta$ , with tan  $\psi$  denoting the amplitude ratio of the two sidebands and  $\theta$  denoting the relative phase of the sidebands in a reference frame rotating at the carrier frequency  $\omega_0$ . Equivalently, as shown mathematically in Eq. (2.8b) and graphically in Fig. 2,  $\theta/2$  gives the azimuth of the semimajor axis of the modulation ellipse generated by the superposition of the two sidebands. The eccentricity of this ellipse is given by

$$e = \frac{2(\sin\psi\cos\psi)^{1/2}}{(\cos\psi + \sin\psi)}, \qquad 0 \le \psi \le \pi/2.$$
(2.9)



Fig. 1. The complex amplitude of the total electric field decomposed into the pump amplitude  $\mathscr{E}_0(z)$  and the probe amplitude  $\delta \tilde{\mathscr{E}}(z, t)$ . In the complex  $\tilde{\mathscr{E}}$  plane,  $\mathscr{E}_0(z)$  lies along the real axis, while  $\delta \tilde{\mathscr{E}}(z, t)$  revolves periodically around an ellipse. For a pure AM probe field, the ellipse degenerates into a straight line along the  $\mathscr{E}'$  axis, while for a pure FM probe field the ellipse degenerates into a straight line parallel to the  $\mathscr{E}''$  axis.



c) superposition  $: \delta \tilde{\epsilon}$ 

Fig. 2. Complex  $\mathscr{E}$ -plane decomposition at fixed z of the probe field as defined in Eq. (2.8). a)  $+\delta\omega$  sideband corresponds to  $\delta\tilde{\mathscr{E}}$  performing a counterclockwise circular rotation at angular frequency  $+\delta\omega$ . b) the  $-\delta\omega$  sideband. c) In general, the total probe field consists of a superposition of  $+\delta\omega$  and  $-\delta\omega$  sidebands, forming an ellipse of eccentricity given by Eq. (2.9) and azimuthal angle  $\theta/2$ .

For the limiting case of a single-sideband probe field,  $\psi$  is equal to 0 or  $\pi/2$ , and in either case the eccentricity *e* is equal to 0, implying that the modulation ellipses are in fact circles. For the case of equal-amplitude sidebands,  $\psi$  is equal to  $\pi/4$ , and *e* is equal to 1, implying that the modulation ellipse is in fact a straight line. The important special cases of pure amplitude and pure frequency modulation are given by

amplitude modulation:  $\psi = \pi/4$ ,  $\theta = 0$ , (2.10a)

frequency modulation:  $\psi = \pi/4$ ,  $\theta = \pi$ . (2.10b)

In general, the parameters  $\psi$  and  $\theta$  depend on the coordinate z, but we have suppressed this dependence because in certain important special cases (the natural modes) these parameters are independent of z.

Consistency between the expansions of the total electric field in terms of its Fourier components Eqs. (2.1)-(2.3)] and in terms of its modulated and carrier-wave components [Eqs. (2.4), (2.5), and (2.8)] is ensured if the field amplitudes are related according to

$$A_0(z) = \mathcal{E}_0(z) \exp[i\phi_0(z)],$$
 (2.11a)

$$A_{-\delta\omega}(z) = \delta \mathscr{E}(z) \sin \psi \exp\{i[\phi_0(z) - \phi_1(z) - \theta]\}, \quad (2.11b)$$

$$A_{+\delta\omega}(z) = \delta \mathscr{E}(z) \cos \psi \exp\{i[\phi_0(z) + \phi_1(z)]\}.$$
(2.11c)

# 3. NATURAL MODES OF MODULATED BEAMS

The nonlinear interaction of the optical field with the material system manifests itself as a coupling among the various components of the total electric field. The coupling between the two weak waves at frequencies  $\omega_0 \pm \delta \omega$  is described by the Helmholtz equations

$$\frac{\partial^2 F_{\pm\delta\omega}}{\partial z^2} + (k_0 \pm \delta k)^2 F_{\pm\delta\omega} = \frac{4\pi (\omega_0 \pm \delta \omega)^2}{c^2} \{\chi^{(1)}(\omega_0 \pm \delta \omega) F_{\pm\delta\omega} + \chi^{(3)}[\omega_0 \pm \delta\omega; \omega_0, \omega_0, -(\omega_0 \mp \delta\omega)] F_0{}^2 F_{\pm\delta\omega} *\}.$$
(3.1)

Here  $\chi^{(1)}$  and  $\chi^{(3)}$  represent first- and third-order susceptibilities generalized to include the possibility of saturation due to the pump field  $F_0$ . Hence  $\chi^{(1)}(\omega_0 \pm \delta \omega)$  gives the effects of saturated absorption and dispersion on the wave at frequency  $\omega_0 \pm \delta \omega$ , whereas  $\chi^{(3)}[\omega_0 \pm \delta \omega; \omega_0, \omega_0, -(\omega_0 \mp \delta \omega)]$ represents the response at frequency  $(\omega_0 \pm \delta \omega)$  due to the wave at frequency  $(\omega_0 \mp \delta \omega)$ . Explicit formulas for  $\chi^{(1)}$  and  $\chi^{(3)}$  for a two-level system have been given elsewhere<sup>4</sup>; formulas for a three-level system are included in Appendix A.

Through introduction of the slowly varying amplitude approximation

$$\left|\frac{\partial^2 A_{\pm\delta\omega}}{\partial z^2}\right| \ll (k_0 \pm \delta k) \left|\frac{\partial A_{\pm\delta\omega}}{\partial z}\right|,$$

the Helmholtz equations reduce to the coupled amplitude equations

$$\frac{\partial}{\partial z}A_{\pm\delta\omega} = -\alpha_{\pm\delta\omega}A_{\pm\delta\omega} + \kappa_{\pm\delta\omega}A_{\pm\delta\omega}^*. \tag{3.2}$$

These two equations can be written conveniently in matrix form as

$$\frac{\partial}{\partial z} \begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^* \end{bmatrix} = \begin{bmatrix} -\alpha_{+\delta\omega} & \kappa_{+\delta\omega} \\ \kappa_{-\delta\omega}^* & -\alpha_{-\delta\omega}^* \end{bmatrix} \begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^* \end{bmatrix}.$$
(3.3)

Here  $\alpha_{\pm\delta\omega}$  and  $\kappa_{\pm\delta\omega}$  represent nonlinear absorption and coupling coefficients, respectively, which are given by

$$\chi_{\pm\delta\omega} = \frac{2\pi i(\omega_0 \pm \delta\omega)}{c} \chi^{(1)}(\omega_0 \pm \delta\omega), \qquad (3.4)$$

$$\kappa_{\pm\delta\omega} = \frac{-2\pi i(\omega_0 \pm \delta\omega)}{c} \chi^{(3)}[\omega_0 \pm \delta\omega; \omega_0, \omega_0, -(\omega_0 \mp \delta\omega)]A_0(z)^2.$$
(3.5)

We note that the linear contribution to the refractive index has been included in  $\alpha$  and that phase-matching effects manifest themselves through the phases of  $\alpha$  and  $\kappa$ .

We now seek solutions of Eq. (3.3) with the special property that the relative amplitudes and phases of  $A_{+\delta\omega}$  and  $A_{-\delta\omega}$  remain unchanged as the wave propagates. These modes hence experience at most an overall complex attenuation  $\lambda$  defined by

$$\frac{\partial}{\partial z} \begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^* \end{bmatrix} = -\lambda \begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^* \end{bmatrix} .$$
(3.6)

We call these solutions natural modes of propagation of the weak field  $E_{+\delta\omega}(z,t) + E_{-\delta\omega}(z,t)$ . By combining Eqs. (3.3) and (3.6), we find that the natural modes are the eigenvectors of the equation

$$\begin{array}{l} \lambda - \alpha_{+\delta\omega} & \kappa_{+\delta\omega} \\ \kappa_{-\delta\omega}^* & \lambda - \alpha_{-\delta\omega}^* \end{array} \Big| \begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^* \end{bmatrix} = 0.$$
 (3.7)

The complex attenuation experienced by one of these modes is given by the eigenvalue  $\lambda$ , which is given by the solution of the secular equation associated with Eq. (3.7) as

$$\lambda_{\pm} = \frac{1}{2} (\alpha_{+\delta\omega} + \alpha_{-\delta\omega}^{*}) \\ \pm \frac{1}{2} [(\alpha_{+\delta\omega} - \alpha_{-\delta\omega}^{*})^{2} + 4\kappa_{+\delta\omega}\kappa_{-\delta\omega}^{*}]^{1/2}.$$
(3.8)

The associated eigenvectors, that is, the natural modes, are given by

$$\mathbf{A}_{+} = \begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^{*} \end{bmatrix}_{+} = N_{+} \begin{bmatrix} 1 \\ B_{+}^{*} \end{bmatrix}$$
(3.9a)

and

$$\mathbf{A}_{-} \equiv \begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^{*} \end{bmatrix}_{-} = N_{-} \begin{bmatrix} B_{-} \\ 1 \end{bmatrix}, \qquad (3.9b)$$

where

$$B_{+} = \frac{\kappa_{-\delta\omega}}{\alpha_{-\delta\omega} - \lambda_{+}^{*}},$$
$$B_{-} = \frac{\kappa_{+\delta\omega}}{\alpha_{+\delta\omega} - \lambda_{-}},$$

and

$$N_{\pm} = (1 + |B_{\pm}|^2)^{-1/2}$$

The significance of Eqs. (3.8) and (3.9) is as follows: If a single-sideband probe field (say,  $A_{+\delta\omega}$ ) is introduced into the partially saturated medium, this wave will be parametrically coupled to the other sideband  $(A_{-\delta\omega})$ . Two linear combinations of these sidebands, which are the natural modes  $(A_{\pm})$ , have the property that they propagate with constant relative amplitude and phase but with overall complex attenuation given by  $\lambda_{\pm}$ . An arbitrary probe-wave input can be decomposed into a linear combination of these two natural modes according to

$$\begin{bmatrix} A_{+\delta\omega} \\ A_{-\delta\omega}^* \end{bmatrix} = c_+ \mathbf{A}_+ + c_- \mathbf{A}_-,$$
(3.10)

where

$$\begin{bmatrix} c_{+} \\ c_{-} \end{bmatrix} = \frac{1}{N_{+}N_{-}(1 - B_{+}*B_{-})} \times \begin{bmatrix} N_{-} & -N_{-}B_{-} \\ -N_{+}B_{+}* & N_{+} \end{bmatrix} \begin{bmatrix} A_{+\delta\omega} \\ A_{-}\delta\omega* \end{bmatrix}.$$
(3.11)

As the probe beam propagates through the saturable absorber it will gradually turn into the natural mode with the lower loss.

## 4. NATURAL MODES FOR THE CASE OF AN ISOLATED ATOMIC RESONANCE

An important special case of the natural modes occurs when the optical nonlinearity is due to the response of a saturable absorber, such as that shown in Fig. 3. For this system, the nonlinear susceptibilities  $\chi^{(1)}$  and  $\chi^{(3)}$ , generalized to include the effects of saturation, are derived in Appendix A. It follows from inspection of Eqs. (A12)–(A19) and Eqs. (3.4) and (3.5) that, for the case of the carrier frequency tuned to exact resonance, the absorption and coupling coefficients are related according to

$$\alpha_{+\delta\omega} = \alpha_{-\delta\omega}^{*},$$
  
$$\kappa_{+\delta\omega} \exp[-2i\phi_0(z)] = \kappa_{-\delta\omega}^{*} \exp[2i\phi_0(z)].$$

Note that, according to Eq. (2.11a),  $A_0 = |A_0| \exp[i\phi_0(z)]$ . The coupled attenuation coefficients are hence given by

$$\lambda_{\pm} = \alpha_{+\delta\omega} \pm (\kappa_{+\delta\omega}\kappa_{-\delta\omega}^*)^{1/2},$$

and the natural modes are given by

$$\mathbf{A}_{+} = \frac{1}{\sqrt{2}} \exp[-i\phi_0(z)] \begin{cases} \exp[i\phi_0(z)] \\ -\exp[-i\phi_0(z)] \end{cases} \rightarrow \mathbf{A}_{\mathrm{FM}}$$

and

$$\mathbf{A}_{-} = \frac{1}{\sqrt{2}} \exp[i\phi_0(z)] \left\{ \exp[i\phi_0(z)] \\ \exp[-i\phi_0(z)] \right\} \rightarrow \mathbf{A}_{\mathrm{AM}}.$$

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That these modes are AM and FM is easily seen by Eqs. (2.10) and (2.11). Hence, for an on-resonant carrier wave, the natural modes of propagation of a modulated beam correspond to pure amplitude and pure frequency modulation.<sup>23,24</sup> The modulated component of a FM wave will always experience attenuation, whereas an AM wave can (if Re  $\lambda < 0$ ) experience amplification.

For the case of an off-resonance pump wave, the natural modes do not in general correspond to pure amplitude and pure frequency modulation. However, amplitude and frequency modulation constitute an appropriate basis set for the natural modes even in this case, since many spectroscopic applications involve the measurement of the AM component of a light wave whose state of modulation has been modified by its interaction with a nonlinear medium. In terms of this basis set, we can derive [analogously to Eq. (3.3)] a coupled amplitude equation for the AM and FM components of the probe field of the form

$$\frac{\partial}{\partial z} \begin{bmatrix} \delta \tilde{\mathcal{E}}_{AM}(z) \\ \delta \tilde{\mathcal{E}}_{FM}(z) \end{bmatrix} = - \begin{bmatrix} \tilde{b}_{AA} & \tilde{b}_{AF} \\ \tilde{b}_{FA} & \tilde{b}_{FF} \end{bmatrix} \begin{bmatrix} \delta \tilde{\mathcal{E}}_{AM}(z) \\ \delta \tilde{\mathcal{E}}_{FM}(z) \end{bmatrix}, \qquad (4.1)$$

where the  $\delta \mathcal{E}_{AM}$  and the  $\delta \mathcal{E}_{FM}$  of Eq. (2.6) are related to the  $\delta \tilde{\mathcal{E}}_{AM}$  and  $\delta \tilde{\mathcal{E}}_{FM}$  of Eq. (4.1) as

$$\delta \mathscr{E}_{AM}(z, t) = \operatorname{Re}\{\delta \mathscr{E}_{AM}(z) \exp[i(\delta \omega t - \delta kz)]\},\$$
  
$$\delta \mathscr{E}_{FM}(z, t) = \operatorname{Re}\{\delta \mathscr{\tilde{E}}_{FM}(z) \exp[i(\delta \omega t - \delta kz - \pi/2)]\}.$$
  
(4.2)

The significance of the matrix elements  $\tilde{b}_{AA}$  and  $\tilde{b}_{FF}$  is that they give the attenuation experienced by the AM or the FM component, respectively. Similarly,  $\tilde{b}_{AF}$  gives the rate at which frequency modulation is converted to amplitude modulation by means of the nonlinearity of the atomic response, whereas  $\tilde{b}_{FA}$  gives the rate at which amplitude modulation is converted to frequency modulation. These coefficients are determined most conveniently by using the Bloch-equation formalism,<sup>25</sup> since this formalism naturally decomposes the atomic polarization into its components in phase with and in quadrature with the driving field. A derivation of these coefficients using this formalism is presented in Appendix B.

In analogy to the case of the single-sideband basis set of Section 3, we define the natural modes to be solutions of Eq. (4.1) having the property that the relative amplitude and phase of the AM and FM sidebands remain fixed as the beam propagates, that is, we require that



Fig. 3. The energy-level depiction of the three-level system is shown. Levels  $|a\rangle$  and  $|b\rangle$ , separated by energy  $\hbar\omega_{ba}$ , are coupled by the strong pump field at  $\omega_0$ . The probe field is at frequency  $\omega_0 \pm \delta\omega$ . Relaxation rates between the levels are represented by the  $\gamma$ 's.

The natural modes hence obey the eigenvalue equation

$$\begin{bmatrix} \tilde{b}_{AA} - \tilde{\lambda} & \tilde{b}_{AF} \\ \tilde{b}_{FA} & \tilde{b}_{FF} - \tilde{\lambda} \end{bmatrix} \begin{bmatrix} \delta \tilde{\mathcal{E}}_{AM} \\ \delta \tilde{\mathcal{E}}_{FM} \end{bmatrix} = 0 \cdot$$
 (4.4)

The eigenvalues, which represent the attenuation of the coupled solution, are given explicity as

$$\lambda_{\pm} = \frac{1}{2} (\tilde{b}_{AA} + \tilde{b}_{FF}) \pm \frac{1}{2} [(\tilde{b}_{AA} - \tilde{b}_{FF})^2 + 4\tilde{b}_{AF} \tilde{b}_{FA}]^{1/2},$$
(4.5)

and the eigenvectors are given by

$$\begin{split} \delta \tilde{\mathscr{E}}_{+} &= \begin{bmatrix} \delta \tilde{\mathscr{E}}_{\mathrm{AM}} \\ \delta \tilde{\mathscr{E}}_{\mathrm{FM}} \end{bmatrix}_{+}^{+} = M_{+} \begin{bmatrix} D_{+} \\ 1 \end{bmatrix}, \\ M_{+} &= \begin{bmatrix} 1 + \left| \frac{\tilde{b}_{\mathrm{FF}} - \lambda_{+}}{\tilde{b}_{\mathrm{FA}}} \right|^{2} \end{bmatrix}^{-1/2}, \\ D_{+} &= \frac{\tilde{b}_{\mathrm{FF}} - \lambda_{+}}{\tilde{b}_{\mathrm{FA}}}, \end{split}$$
(4.6a)

and

$$\begin{split} \delta \tilde{\mathscr{E}}_{-} &= \begin{bmatrix} \delta \tilde{\mathscr{E}}_{\mathrm{AM}} \\ \delta \tilde{\mathscr{E}}_{\mathrm{FM}} \end{bmatrix}_{-} = M_{-} \begin{bmatrix} 1 \\ D_{-} \end{bmatrix}, \\ M_{-} &= \begin{bmatrix} 1 + \left| \frac{\delta_{\mathrm{AA}} - \lambda_{-}}{\delta_{\mathrm{AF}}} \right|^{2} \end{bmatrix}^{-1/2}, \\ D_{-} &= \frac{\delta_{\mathrm{AA}} - \lambda_{-}}{\delta_{\mathrm{AF}}}. \end{split}$$
(4.6b)

Any arbitrary input  $\delta \tilde{\mathcal{E}}$  can be decomposed into a linear combination of these natural modes as

$$\delta \tilde{\mathscr{E}} = \begin{bmatrix} \delta \tilde{\mathscr{E}}_{\mathrm{AM}} \\ \delta \tilde{\mathscr{E}}_{\mathrm{FM}} \end{bmatrix} = c_{+} \delta \tilde{\mathscr{E}}_{+} + c_{-} \delta \tilde{\mathscr{E}}_{-}, \qquad (4.7)$$

where the decomposition coefficients are given by

$$\begin{bmatrix} c_{-} \\ c_{+} \end{bmatrix} = \frac{1}{M_{+}M_{-}(1-D_{+}D_{-})} \begin{bmatrix} M_{-}D_{-} & -M_{-} \\ -M_{+} & M_{+}D_{+} \end{bmatrix} \begin{bmatrix} \delta \tilde{e}_{AM} \\ \delta \tilde{e}_{FM} \end{bmatrix} .$$
(4.8)

#### 5. EXAMPLES OF NATURAL-MODE SOLUTIONS FOR AN ISOLATED ATOMIC RESONANCE

In order to give insight into the nature of the natural modes, we present in this section graphical depictions of the natural-mode solution for several representative cases. These cases are identified by the ratio  $(T_1/T_2)$  of the population to dipole relaxation times, by the detuning  $(\Delta = \omega_0 - \omega_{ba})$  of the carrier frequency  $\omega_0$  from the atomic-resonance frequency  $\omega_{ba}$ , of the Rabi frequency  $\Omega$  introduced in Eq. (A11), and of the dimensionless intensity  $I_0 = \Omega^2 T_1 T_2$ . Graphical depictions of the natural-mode solutions [Eqs. (4.5) and (4.6)] to the eigenvalue equation [Eq. (4.4)] are shown in Figs. 4–6. In each of Figs. 4-6, the upper two rows show the natural modes given by Eq. (4.6) in terms of the parameters  $\theta$  and sin  $\psi$  as defined in Eqs. (2.8). The lower two rows show the fieldabsorption and -dispersion coefficients for the natural modes obtained from the complex eigenvalue given in Eq. (4.5). These curves are normalized such that the on-resonance unsaturated field-absorption coefficient is unity. Figure 4 illustrates the weak-field ( $I_0 \ll 1)$  limit for the case of radiative broadening (i.e.,  $T_1/T_2 = 0.5$ ) for two different values of the

carrier-wave detuning. We note that when the pump frequency is equal to that of the atomic resonance (i.e.,  $\Delta = 0$ ), the natural modes are pure AM and FM fields. This is true whenever  $\Delta = 0$  irrespective of pump intensity, probe detuning, or the presence of collisional dephasing. It is also seen from Fig. 4 that for small modulation frequencies  $||\delta\omega| \leq (\Omega^2)$  $(+ \Delta^2)^{1/2}$  the AM field always experiences less absorption than the FM field. The AM component of any general modulation form will under these conditions dominate the output after propagation through sufficient length of a noninverted medium. For larger modulation frequencies, a FM wave can experience less attenuation than an AM wave.<sup>24</sup> and the FM component will hence dominate. When the pump is detuned from the atomic resonance (see the second column of Fig. 4), the natural modes are no longer pure amplitude and frequency modulation forms but some linear combination of the two. The minus mode (which has the larger AM component) still experiences less absorption for small modulation frequencies than the plus mode.

$$I_0 = 0.2$$
  $I_0 = 0.2$   
 $\Delta T_2 = 0.0$   $\Delta T_2 = 1.0$   
 $T_1 / T_2 = 0.5$   $T_1 / T_0 = 0.5$ 



 $\delta \omega T_2$ : modulation frequency

Fig. 4. The natural-mode solution is illustrated for the case of radiative broadening  $(T_1/T_2 = 0.5)$  for  $I_0 = 0.2$ . The solid lines represent the minus mode, while the dashed lines represent the plus mode. The two cases illustrated are (left)  $\Delta T_2 = 0$  and (right)  $\Delta T_2 = 1$ .



Fig. 5. The natural-mode solution is illustrated for the case of rapid collisional dephasing  $(T_1/T_2 = 50)$  for  $I_0 = 0.2$ . The solid lines represent the minus mode, while the dashed lines represent the plus mode. Illustrated are (left)  $\Delta T_2 = 0$ , (center and right)  $\Delta T_2 = 1$ . The right-hand column illustrates the solution for small modulation frequencies.

Figure 5 illustrates the weak-field limit for the case of rapid collisional dephasing  $(T_1/T_2 = 50)$ . The leftmost column refers to an on-resonance carrier frequency, whereas the other two columns refer to a carrier wave that is detuned by one atomic linewidth. These columns differ in that the rightmost column shows the abscissa with a much expanded scale. For both detunings, a narrow spectral hole of width  $1/T_1$  is present in the absorption profile of the minus mode. Experimental observations of this feature have been made and are discussed in Section 7.

Figure 6 shows the natural-mode solution for the case in which the Rabi frequency  $\Omega$  associated with the atomic response is greater than both the detuning  $\Delta$  and the atomic linewidth  $1/T_1$ . The absorptive response for the plus mode shows a resonance near the generalized Rabi frequency  $(\Omega^2 + \Delta^2)^{1/2}$ . For the minus mode, a broad region of negative absorption occurs, extending from zero modulation frequency to approximately the generalized Rabi frequency. As a result, radiation near the Rabi sidebands can be generated spontaneously by growing from noise within the nonlinear medium, as was recently observed experimentally.<sup>26</sup> This effect can also lead to instabilities in homogeneously broadened lasers, as was also observed recently.<sup>27</sup>

The form of the natural modes has been illustrated in Figs.

4-6 by plotting the relative sideband amplitude  $\sin \psi$  and sideband phase difference  $\theta$  [see Eqs. (2.8)] as functions of the modulation frequency. Alternatively, the form of the natural modes can be illustrated by displaying the form of the modulation ellipse, as introduced in the phase-plane diagram of Fig. 2. The modulation forms for the specific cases discussed above are shown in Fig. 7. For the case of a resonant carrier wave (i.e.,  $\Delta T_2 = 0$ ), the ellipses collapse into horizontal and vertical straight lines (corresponding to amplitude and frequency modulation, respectively) for any value of  $T_1/T_2$ , of  $I_0$ , and of  $\delta\omega$ . In the remainder of the examples,  $\Delta T_2$  is not equal to 0, and the form of the natural modes depends on the modulation frequency. In all such cases, the modulation ellipses approach circles for large modulation frequencies, because at large modulation frequencies the two sidebands are only weakly coupled and hence propagate essentially independently. At low modulation frequencies, the ellipses approach straight lines, because the two sidebands are strongly coupled in this case, and the natural modes consist of roughly



Fig. 6. The natural-mode solution is illustrated for the strong pump field limit. The solid lines represent the minus mode, and the dashed lines represent the plus mode. Illustrated are (left)  $\Delta T_2 = 3$ ,  $\Omega T_2 = 8$ , and  $T_1/T_2 = 0.5$ ; (right)  $\Delta T_2 = 3$ ,  $\Omega T_2 = 8$ , and  $T_1/T_2 = 50$ .



Fig. 7. The modulation forms for the complex electric field of the natural modes are shown. The vertical axis corresponds to the FM component, and the horizontal axis corresponds to the AM component as shown in Figs. 1 and 2.

equal amounts of the two sidebands. These figures show that the details of how the modes evolve with increasing modulation frequency depend on the carrier-wave intensity and detuning and on the ratio  $T_1/T_2$ .

### 6. MODULATION SPECTROSCOPY

Conventional FM saturation spectroscopy is accomplished by sending a weakly modulated FM wave through a nonlinear medium and measuring the in-phase and in-quadrature components of the AM output. A spectrum of the material can be obtained by scanning the frequency of the laser at constant modulation frequency or by scanning the modulation frequency at constant laser frequency. The spectral line shapes obtained with FM saturation spectroscopy are easily found from the natural-mode formalism. Since a square-law detector is sensitive only to amplitude modulation, the signal S(t) from such a detector is proportional (for  $\delta E \ll E_0$ ) to

$$S(t) \propto \mathcal{E}_0^2 + 2\mathcal{E}_0 \operatorname{Re}[\delta \tilde{\mathcal{E}}_{AM} \exp(i\delta\omega t)].$$
(6.1)

If a purely FM probe field given by

$$\delta \mathcal{E}_{\rm FM}(z=0) = M \mathcal{E}_0(z=0)$$

(where the modulation index M is assumed to be much less than unity) is sent through an optically thin medium of length l, the complex amplitude of the emerging AM component is

$$\delta \tilde{\mathcal{E}}_{AM}(l) = -l \tilde{b}_{AF} \delta \tilde{\mathcal{E}}_{FM}(0). \tag{6.2}$$

The expression [relation (6.1)] for the signal produced by the square-law detector is then given by

$$S(t) \propto E_0^2 - 2lM \mathcal{E}_0^2 (\operatorname{Im} \tilde{b}_{AF} \cos \delta \omega t + \operatorname{Re} \tilde{b}_{AF} \sin \delta \omega t).$$
(6.3)

A lock-in amplifier may then be used to separate the in-phase and in-quadrature components of the signal. In Figs. 8 and



Fig. 8. The in-phase (top) and in-quadrature (bottom) FM saturation-spectroscopy spectrum as a function of the laser detuning  $\Delta T_2$ . The modulation frequency is fixed at  $\delta\omega T_2 = 8$ , and  $T_1/T_2 = 0.5$ . The three curves correspond to Rabi frequencies of  $\kappa T_2 \mathcal{E}_0 = 0$ , 4, and 8, which correspond to intensities of  $I_0 = 0$ , 8, and 32, respectively.



Fig. 9. The in-phase (top) and in-quadrature (bottom) FM saturation-spectroscopy spectrum as a function of the modulation frequency  $\delta\omega T_2$ . The laser detuning is fixed at  $\Delta T_2 = 8$ , and  $T_1/T_2 = 0.5$ . The three curves correspond to Rabi frequencies of  $\kappa T_2 \mathcal{E}_0 = 0$ , 4, and 8, which correspond to  $I_0 = 0$ , 8, and 32, respectively.

9, examples of possible outputs from such a modulationspectroscopy experiment are shown. These results are general in that they include both the linear response and the nonlinear response of the media. The signal-to-noise merits of such techniques are well established.<sup>18,20</sup>

#### 7. EXPERIMENTAL EXAMPLES OF AMPLITUDE-MODULATION SATURATION SPECTROSCOPY

In this section, we describe the results of several spectroscopic studies conducted using AM saturation spectroscopy, a spectroscopic technique in which the change in the depth of modulation of an AM beam is measured. These experiments are typically carried out at or near resonance, for which case amplitude modulation is a natural mode of propagation. The relation between the output and input fields is thus given by the  $\delta_{AA}$  matrix element of Eq. (4.1) or (4.4).

AM saturation spectroscopy provides a sensitive tool for determining population-relaxation processes in nonlinear optical materials. It was pointed out by Schwartz and Tan<sup>28</sup> in 1967 that a spectral hole of width  $T_1^{-1}$  (where  $T_1$  here represents the ground-state recovery time) occurs in the probe absorption profile of a homogeneously broadened saturable absorber in the presence of a saturating pump wave. Just as in spectral hole burning in inhomogeneously broadened media, this hole occurs at the pump frequency. The origin of this spectral feature can be traced to the oscillation in the ground-state population at the beat frequency between the pump and probe waves.<sup>29</sup> The population is able to respond only if the beat frequency is comparable to or less than  $1/T_1$ . The material therefore becomes a temporally modulated absorber, and this effect increases the degree of modulation of the transmitted beam. A simple rate-equation treatment of the phenomenon is presented in Appendix C. Of course, this effect is also properly described by the more general theory developed in Sections 2 and 3 of this paper (see Fig. 5).

Figure 10 shows a series of AM spectra for ruby<sup>30</sup> for several different pump intensities. At low intensities, a spectral hole of half-width 43 Hz ( $\sim 1/T_1$ ) is observed. At higher pump intensities, the spectral hole is broader, because the strong pump field actively increases the rate of relaxation of the system. The data are in good agreement with the solid theoretical curves that are calculated using the natural-mode formalism.

Qualitatively different behavior is observed when AM spectroscopy is performed on alexandrite. In this material, excited-state absorption plays an important role in determining the nature of the atomic saturation. For wavelengths between  $\sim$ 450 and 500 nm, the absorption cross section of the excited state is larger than that of the ground state. At these wavelengths alexandrite acts as an inverse saturable absorber in that the absorption coefficient of the material increases (rather than decreases) with increasing laser intensity. As a result, a spectral antihole rather than a hole is observed when AM spectroscopy is performed at these wavelengths.<sup>31</sup> Figure 11 shows the AM spectrum of alexandrite at a wavelength of 457 nm where such an antihole appears.

For ruby and alexandrite, the spectral hole is found to have a Lorentzian line shape at low laser intensities. Non-Lorentzian line shapes are observed for the case of fluorescein in boric acid glass. For this material, following optical excitation from the singlet ground state to the singlet excited state, an intersystem crossing can occur, leading to the trapping of population in the long-lived triplet excited state.<sup>32</sup> At room temperature, the dominant decay route out of this state is a thermally activated transfer back to the singlet excited state followed by a radiative decay to the ground state. This process is known as delayed fluorescence. For fluorescein in boric acid glass, this process leads to a nonexponential luminescent decay in times of the order of 1 sec. It is believed that the reason why this decay is nonexponential is that the thermally activated transfer out of the triplet state occurs at different rates for different sites within the glass matrix.<sup>33</sup> Owing to the nonexponential character of the population decay, the



Fig. 10. AM-spectroscopy results for ruby. The attenuation of the AM component is shown as a function of modulation frequency for several different degrees of saturation. Note the existance of a spectral hole at low modulation frequencies, as discussed in the text.



Fig. 11. AM spectrum for alexandrite at a laser wavelength of 457 nm. At this wavelength alexandrite shows inverse saturation, leading to the observed antihole.



Fig. 12. AM spectrum for fluorescein in glass. This material shows nonexponential population decay, leading to the observed markedly non-Lorentzian spectrum. Also shown for component is the upper curve, which is a Lorentzian that is constrained to fit well in the wings.

spectral hole observed by AM spectroscopy has the markedly cusped, non-Lorentzian shape shown in Fig. 12. The width of this feature is 750 mHz (HWHM). These examples show that a wealth of information regarding population-decay dynamics is contained in the AM spectrum.

#### CONCLUSIONS

We have shown that there exist two natural modes of propagation of a weakly modulated beam propagating through any nonlinear optical medium. These natural modes have the property that the form of modulation remains invariant as the beam propagates. If a modulation form other than one of these natural modes is injected into the medium, the type of modulation is transformed on propagation into the natural mode having the lower attenuation. Formulas for these natural modes have been presented both for the case of a general optical nonlinearity and for the special case of a medium that can be modeled as a collection of two-level atoms. The natural-mode formalism provides a basis for treating all types of modulation spectroscopy involving weakly modulated beams. This theory permits the treatment of both frequencyand amplitude-modulation spectroscopy. These two spectroscopies are complementary in their regimes of usefulness. FM saturation spectroscopy has been used to locate spectral lines and measure dipole dephasing rates with great accuracy. Conversely, AM saturation spectroscopy is rather insensitive to the location of the line center but provides a sensitive probe of population-relaxation processes.

#### APPENDIX A

In this appendix, we calculate the response of the three-level system shown in Fig. 3 to an applied optical field E. We assume that this field can interact with levels a and b and that relaxation through level c is a possible decay route. The equations describing the time evolution of the density matrix for this system are

$$\dot{\rho}_{aa} = \gamma_{ba}\rho_{bb} + \gamma_{ca}\rho_{cc} + \frac{1}{i\hbar}\left(\rho_{ab}\mu_{ba}E - \mu_{ab}E\rho_{ba}\right), \qquad (A1)$$

$$\dot{\rho}_{bb} = -\gamma_{ba}\rho_{bb} - \gamma_{bc}\rho_{bb} + \frac{1}{i\hbar}(\rho_{ba}\mu_{ab}E - \mu_{ba}E\rho_{ab}), \quad (A2)$$

$$\dot{\rho}_{cc} = \gamma_{bc} \rho_{bb} - \gamma_{ca} \rho_{cc}, \tag{A3}$$

$$\dot{\rho}_{ba} = -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba} + \frac{1}{i\hbar}\,\mu_{ba}E(\rho_{bb} - \rho_{aa}).\tag{A4}$$

Here  $T_2$  represents the dipole relaxation time and  $\gamma_{ij}$  represents the relaxation rate between levels *i* and *j*.

We assume for simplicity that the total applied field can be represented as the sum of two Fourier components by

$$E = F_0 \exp(i\omega_0 t) + F_{\pm\delta\omega} \exp[i(\omega_0 \pm \delta\omega)t], \quad (A5)$$

where  $F_0$  and  $F_{\pm\delta\omega}$  represent the amplitudes of the pump and probe fields, respectively. The probe field is considered to be sufficiently weak that we require solutions to Eqs. (A1)– (A4) that are correct only to lowest order in  $F_{\pm\delta\omega}$ . The steady-state solutions to these equations can be obtained in the rotating-wave approximation in terms of the Fourier amplitudes of the density-matrix elements as

$$i\delta\omega\rho_{aa}(-\delta\omega) = \gamma_{ba}\rho_{bb}(-\delta\omega) + \gamma_{ca}\rho_{cc}(-\delta\omega) + \frac{1}{i\hbar} \left[\rho_{ab}(-\omega_0 - \delta\omega)\mu_{ba}F_0 - \rho_{ba}(\omega_0)\mu_{ab}F_{+\delta\omega}* - \rho_{ba}(\omega_0 - \delta\omega; \omega_0, \omega_0, \omega_0 - \delta\omega)\mu_{ab}F_0*\right],$$
(A6)

$$\begin{split} i\delta\omega\rho_{bb}(-\delta\omega) &= -(\gamma_{ba} + \gamma_{bc})\rho_{bb}(-\delta\omega) \\ &- \frac{1}{i\hbar} \left[ \rho_{ab}(-\omega_0 - \delta\omega)\mu_{ba}F_0 \right. \\ &+ \rho_{ba}(\omega_0)\mu_{ab}F_{+\delta\omega}* \\ &- \rho_{ba}(\omega_0 - \delta\omega; \omega_0, \omega_0, -\omega_0 - \delta\omega)\mu_{ab}F_0* \right], \end{split}$$

$$\rho_{cc}(-\delta\omega) = \frac{\gamma_{bc}\rho_{bb}(-\delta\omega)}{(i\delta\omega + \gamma_{ca})},\tag{A8}$$

$$\rho_{ba}(\omega_0) = \frac{\mu_{ba} F_0(\rho_{bb} - \rho_{aa})^{dc}}{\hbar(\omega_0 - \omega_{ba} + i/T_2)},\tag{A9}$$

$$-i\omega_{3}\rho_{ba}(\omega_{0}+\delta\omega)=-(i\omega_{ba}+1/T_{2})\rho_{ba}(\omega_{0}+\delta\omega)$$

$$+\frac{1}{i\hbar}(\rho_{bb}-\rho_{aa})^{dc}\mu_{ba}F+\delta\omega$$
$$+\frac{1}{i\hbar}\mu_{ba}F_0[\rho_{bb}(\delta\omega)-\rho_{aa}(\delta\omega)],$$

(A10)

and

$$(\rho_{bb} - \rho_{aa})^{dc} = \frac{[1 + (\omega_0 - \omega_{ba})^2 T_2^2](\rho_{bb} - \rho_{aa})^{eq}}{\left[1 + (\omega_0 - \omega_{ba})^2 T_2^2 + \frac{(2 + \gamma_{bc}/\gamma_{ca})}{2(\gamma_{ba} + \gamma_{bc})} T_2\Omega^2\right]},$$
(A11)

where  $\Omega^2 = 4|\mu|^2|F|^2/\hbar^2$  and  $(\rho_{bb} - \rho_{aa})^{eq}$  is the equilibrium value of the population inversion. Equations (A5) and (A6) may be related to yield

$$\rho_{aa}(-\delta\omega) = -\left[1 + \frac{\gamma_{bc}}{(i\delta\omega + \gamma_{ca})}\right]\rho_{bb}(-\delta\omega)$$
$$= -f^*\rho_{bb}(-\delta\omega). \tag{A12}$$

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Algebraic manipulation then gives the following solutions:

$$\rho_{bb}(\delta\omega) = \frac{1}{\hbar} \frac{(\delta\omega + 2i/T_2)(\omega_0 - \delta\omega - \omega_{ba} - i/T_2)(\rho_{bb} - \rho_{aa})^{dc} |\mu|^2 F_0 * F_{+\delta\omega}}{(\omega_0 - \omega_{ba} + i/T_2)D(\omega_0 + \delta\omega)},$$
(A13)

$$\rho_{ba}(\omega_0 + \delta\omega) = \frac{\mu_{ba}F_{+\delta\omega}(\rho_{bb} - \rho_{aa})^{dc}}{D(\omega_0 + \delta\omega)} \left\{ [\delta\omega + i(\gamma_{ba} + \gamma_{bc})](\delta\omega - \omega_0 + \omega_{ba} + i/T_2) - \frac{\Omega^2(1+f)\delta\omega}{4(\omega_0 - \omega_{ba} - i/T_2)} \right\}, \quad (A14)$$

$$\rho_{ab}(\delta\omega - \omega_0; -\omega_0, -\omega_0 + \delta\omega) = \frac{F_0 * {}^2F_{+\delta\omega}\mu_{ab}|\mu|^2(\rho_{bb} - \rho_{aa})^{dc}(1+f)(\delta\omega + 2i/T_2)}{\hbar^3(\omega_0 - \omega_{ba} - i/T_2)D(\omega_0 + \delta\omega)},\tag{A15}$$

with

 $D(\omega_0 + \delta\omega) = [\delta\omega + i(\gamma_{ba} + \gamma_{bc})](\delta\omega - \omega_0 + \omega_{ba} + i/T_2)$  $\times (\delta\omega + \omega_0 - \omega_{ba} + i/T_2)$ 

$$-\frac{\Omega^2}{2} (1+f)(\delta\omega + i/T_2).$$
 (A16)

This solution reduces to that obtained for a pure two-level<sup>4</sup> atom by taking the limit  $\gamma_{bc} = 0$ , in which case f = 1.

We can now obtain explicit formulas for the nonlinear polarization in terms of these density-matrix solutions. The nonlinear polarization at frequency  $\omega_0 \pm \delta \omega$  can be written as

$$\mathcal{P}_{\omega_0 \pm \delta \omega}(z, t) = P_{\omega_0 \pm \delta \omega} \exp[i(\omega_0 \pm \delta \omega)t] + \text{c.c.},$$

where the polarization amplitudes are given by

$$P_{\omega_0 \pm \delta \omega} = \chi^{(1)}(\omega_0 \pm \delta \omega) F_{\pm \delta \omega} + \chi^{(3)}(\omega_0 \pm \delta \omega; \omega_0, \omega_0, -\omega_0 \pm \delta \omega) F_0^2 F_{\omega_0 \mp \delta \omega}.$$
(A17)

Neglecting local-field contributions, the generalized first- and third-order susceptibilities are hence given by

$$\chi^{(1)}(\omega_0 \pm \delta \omega) = -N\mu_{ab}\rho_{ba}(\omega_0 \pm \delta \omega)/F_{\omega_0 \pm \delta \omega}, \quad (A18)$$

$$\begin{split} \chi^{(3)}(\omega_0 \pm \delta\omega; \omega_0, \omega_0, -\omega_0 \pm \delta\omega) &= -N\mu_{ab}\rho_{ba}(\omega_0 \pm \delta\omega; \omega_0, \omega_0, \\ &-\omega_0 \pm \delta\omega)/F_0{}^2F_{\omega_0 \pm \delta\omega}^*. \end{split} \tag{A19}$$

#### APPENDIX B

In this appendix, we derive explicit forms for the coefficients that appear in Eq. (4.1). These coefficients are the absorption and gain coefficients for the AM and FM portions of the modulated field. We describe the atomic response in terms of the Bloch vector (u, v, w), whose components are related to the elements of the density matrix by

$$u + iv = 2\rho_{ba} \exp(-i\omega_0 t), \tag{B1a}$$

$$w = \rho_{bb} - \rho_{aa}.$$
 (B1b)

We write the Block equations in matrix form as

$$\frac{\mathrm{d}}{\mathrm{d}t} \begin{bmatrix} u \\ v \\ w \end{bmatrix} = \begin{bmatrix} -1/T_2 & -\Delta & -\kappa \mathcal{E}'' \\ \Delta & -1/T_2 & \kappa \mathcal{E}' \\ \kappa \mathcal{E}'' & -\kappa \mathcal{E}' & -1/T_1 \end{bmatrix} \begin{bmatrix} u \\ v \\ w \end{bmatrix} + \begin{bmatrix} 0 \\ 0 \\ w_{\mathrm{eq}}/T_1 \end{bmatrix}.$$
(B2)

where  $\mathscr{E}'$  and  $\mathscr{E}''$  denote the real and imaginary parts of  $\widetilde{\mathscr{E}}(z, t)$  of Eq. (2.4),  $\kappa = 2|\mu_{ab}|/\hbar$ ,  $\Delta = \omega_{ab} - \omega_0$ ,  $T_1$  and  $T_2$  denote the longitudinal and transverse decay times, and  $w_{eq}$  denotes the equilibrium population difference in the absence of the applied fields. For the case in which the electric-field am-

plitude  $\tilde{\mathscr{E}}(z, t)$  can be decomposed into a pump component  $\mathscr{E}_0(z)$  and a weak probe component  $\delta \tilde{\mathscr{E}}(z, t)$ , the Bloch vector can also be decomposed as

$$\begin{bmatrix} u \\ v \\ w \end{bmatrix} = \begin{bmatrix} u_0 \\ v_0 \\ w_0 \end{bmatrix} + \begin{bmatrix} \delta u \\ \delta v \\ \delta w \end{bmatrix}, \quad (B3)$$

where the first term is the response that is due to the pump field alone and the second term gives the change in the response that is linear in the amplitude of the probe field. For a continuous-wave pump field  $\mathcal{E}_0(z)$ , the steady-state response of Eq. (4.4) is<sup>25</sup>

$$u_0(z) = -w_{\rm eq} \frac{(\Delta T_2)\kappa T_2 \mathcal{E}_0(z)}{[1 + (\Delta T_2)^2 + I_0(z)]},$$
 (B4a)

$$w_0(z) = w_{\rm eq} \frac{\kappa T_2 \mathcal{E}_0(z)}{\left[1 + (\Delta T_2)^2 + I_0(z)\right]},$$
 (B4b)

and

$$w_0(z) = w_{\rm eq} \frac{1 + (\Delta T_2)^2}{[1 + (\Delta T_2)^2 + I_0(z)]},$$
 (B4c)

where we have introduced the dimensionless intensity

$$I_0(z) = \kappa^2 T_1 T_2 \mathcal{E}_0^2(z).$$
(B5)

The equation of motion for the change in the Bloch vector that is due to the perturbing field  $\delta \tilde{\mathscr{E}}$  is

$$\frac{\mathrm{d}}{\mathrm{d}t} \begin{bmatrix} \delta u \\ \delta v \\ \delta w \end{bmatrix} = \begin{bmatrix} -1/T_2 & -\Delta & 0 \\ \Delta & -1/T_2 & \kappa \mathcal{E}_0 \\ 0 & -\kappa \mathcal{E}_0 & -1/T_1 \end{bmatrix} \begin{bmatrix} \delta u \\ \delta v \\ \delta w \end{bmatrix} + \begin{bmatrix} -\kappa w_0 \delta \mathcal{E}_{\mathrm{FM}} \\ \kappa w_0 \delta \mathcal{E}_{\mathrm{AM}} \\ \kappa u_0 \delta \mathcal{E}_{\mathrm{FM}} - \kappa v_0 \delta \mathcal{E}_{\mathrm{AM}} \end{bmatrix} .$$
(B6)

This equation is solved to obtain  $\delta u$  and  $\delta v$  in terms of  $\delta \mathscr{E}_{AM}$ and  $\delta \mathscr{E}_{FM}$ . Since  $\delta u$  and  $\delta v$  represent the contributions to the induced dipole moment, they act as source terms to the inhomogeneous wave equation, which can be written in the slowly varying amplitude approximation as

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right) \begin{bmatrix} \delta \mathscr{E}_{AM} \\ \delta \mathscr{E}_{FM} \end{bmatrix} = \frac{\partial \phi_0(z)}{\partial z} \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix} \begin{bmatrix} \delta \mathscr{E}_{AM} \\ \delta \mathscr{E}_{FM} \end{bmatrix} + \frac{\alpha}{2\kappa T} \begin{bmatrix} \delta \upsilon \\ -\delta u \end{bmatrix},$$
(B7)

where  $\alpha$  is the weak-field line-center absorption coefficient given by

$$\alpha = 4\pi N |\mu_{ab}|^2 \omega T_2 / \hbar c. \tag{B8}$$

By means of a straightforward but lengthy calculation, the Block-vector components  $\delta u$  and  $\delta v$  can be eliminated through use of Eq. (B6), yielding an equation of the form of Eq. (41), with matrix elements given by

$$\tilde{b}_{AA} = \frac{\alpha_0}{2\Gamma} (1 + i\delta\omega T_2) \left[ 1 + (\Delta T_2)^2 - \frac{I_0(z)}{(1 + i\delta\omega T_1)} \right], \quad (B9a)$$

$$\tilde{b}_{\rm AF} = \frac{\alpha_0}{2\Gamma} \Delta T_2^2 \delta \omega (2 + i \delta \omega T_2), \tag{B9b}$$

$$\tilde{b}_{\rm FA} = \frac{\alpha_0}{2\Gamma} \Delta T_2 (2 + i\delta\omega T_2) \left[ \delta\omega T_2 - \frac{I_0(z)}{(1 + i\delta\omega T_1)} \right], \qquad (B9c)$$

$$\tilde{b}_{\rm FF} = \frac{\alpha_0}{2\Gamma} \left\{ (1 + i\delta\omega T_2) [1 + (\Delta T_2)^2] + \frac{I_0(z)}{(1 + i\delta\omega T_1)} \right\}, \quad (B9d)$$

where

$$\Gamma = (1 + i\delta\omega T_2)^2 + (\Delta T_2)^2 + \left[\frac{1 + i\delta\omega T_2}{(1 + i\delta\omega T_1)}\right]I_0(z), \quad (B10)$$

and where the saturated pump absorption coefficient is given by

$$\alpha_0 = -w_{\rm eq} \frac{\alpha}{1 + (\Delta T_2)^2 + I_0(z)}.$$
 (B11)

#### APPENDIX C

In this appendix, it is shown using the rate-equation approximation that a spectral hole that is due to population oscillations having a width  $1/T_1$  is present in the probe absorption profile. In the rate-equation limit, the equation of motion for the ground-state population n is

$$\frac{\mathrm{d}n(t)}{\mathrm{d}t} = -\frac{n\,\sigma I(t)}{\hbar\omega} + \frac{\overline{n} - n(t)}{T_1},\tag{C1}$$

where  $\sigma$  is the absorption cross section,  $\overline{n}$  is the total population, and I is the total intensity, which may be time varying. For the case of a beam of constant intensity  $I_0$ , the groundstate population is given by the steady-state solution of Eq. (C1) as

$$n = \overline{n}/(1 + I_0/I_s), \tag{C2}$$

where the saturation intensity is defined by

$$I_s = \hbar \omega / \sigma T_1. \tag{C3}$$

Note that  $I_0$  in this appendix is a true intensity, not a dimensionless intensity as in Eq. (B5). For the case of a weakly intensity modulated beam, I(t) is given by

$$I = I_0 + [I(\delta\omega)\exp(-i\delta\omega t) + c.c], \qquad (C4)$$

with  $|I(\delta\omega)| \ll I_0$ . We seek solutions of the form

$$n = n_0 + [n(\delta\omega)\exp(-i\delta\omega t) + \text{c.c.}], \quad (C5)$$

with  $|n(\delta \omega)| \ll n_0$ . On substitution of Eq. (C4) and (C5) into Eq. (C1) we obtain

$$n(\delta\omega) = -\frac{\overline{n}}{(1+I_0/I_s)} \frac{(1+I_0/I_s+i\delta\omega T_1)I(\delta\omega)/I_s}{[(1+I_0/I_s)^2+(\delta\omega T_1)^2]}.$$
(C6)

The attenuation of the intensity that is due to propagation through the medium is given by Kramer et al.

$$\frac{\mathrm{d}I(t)}{\mathrm{d}z} = -n(t)I(t). \tag{C7}$$

Separating the Four components of this equation gives

$$\frac{\mathrm{d}}{\mathrm{d}z}I(\delta\omega) = -\sigma n_0 I(\delta\omega) - \sigma n(\delta\omega)I_0$$

$$= -\left(\frac{\sigma \overline{n}}{1 + I_0/I_s}\right)I(\delta\omega)$$

$$+ \left\{\frac{\sigma \overline{n}(1 + I_0/I_s + i\delta\omega T_1)I(\delta\omega)/I_s}{(1 + I_0/I_s)[(1 + I_0/I_s)^2 + (\delta\omega T_1)^2]}\right\}I_0. \quad (C8)$$

The first term is seen to be the normal saturated absorption acting on the modulated component of the intensity. The second term is a gain term for the modulated intensity caused by the temporally modulated absorption acting on the dc portion of the intensity. The origin of the spectral hole is the second term of Eq. (C8). An absorption coefficient for the modulated component of the intensity may be defined as

$$\begin{aligned} \alpha(\delta\omega) &= -\frac{1}{I(\delta\omega)} \frac{\mathrm{d}}{\mathrm{d}z} I(\delta\omega) \\ &= -\frac{\sigma\overline{n}}{(1+I_0/I_s)} \left[ 1 - \frac{(1+I_0/I_s+i\delta\omega T_1)I_0/I_s}{(1+I_0/I_s)^2 + (\delta\omega T_1)^2} \right], \quad (C9) \end{aligned}$$

showing explicitly the spectral hole of half-width  $1/T_1$  at low values of  $I_0$ .

#### ACKNOWLEDGMENTS

We gratefully acknowledge helpful discussions with A. Szabo and our associates J. Krasinski, M. S. Malcuit, and M. G. Raymer. This research was supported by the National Science Foundation through grant ECS-811821 and by the Joint Services Optics Program.

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