Theory of a Zeeman ring laser: General formalism

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A theory of a ring laser subject to a uniform, axial dc magnetic field is given in extension of the two-mirror standing-wave treatment by Sargent, Lamb, and Fork. The active medium consists of thermally moving atoms that have two electronic levels with arbitrary angular momenta. The electric field is treated classically for two circular polarizations of opposite sense in a cavity with any degree of cavity anisotropy. Losses due to backscattering are also included. In addition, the results of a generalized treatment are given which includes arbitrarily oriented magnetic field, a general state of electric field polarization, varying isotopic abundance, and hyperfine structure. The self-consistency requirement is used to obtain amplitude- and frequency-determining equations for multimode operation as functions of laser parameters. A general calculational technique, the "perturbation tree," is introduced in the calculation of the third-order component of the population matrix, greatly simplifying the algebra involved by allowing it to be abstracted in tabular form.

I. INTRODUCTION

In a paper by Sargent, Lamb, and Fork,[1] the multimode theory of an optical maser (henceforth referred to as the scalar theory) developed by Lamb[2] was extended to include general states of electric field polarization and cavity frequency and loss anisotropy, a dc magnetic field at arbitrary angles to the maser axis, and an active medium consisting of two-level atoms (arbitrary number of isotopes) having two electronic states, each of which could have arbitrary angular momentum and hyperfine structure. That theory was given for a standing-wave two-mirror laser configuration.

In the present paper we consider a ring-laser configuration diagrammed in Fig. 1. The electric field is represented by a set of traveling-wave "modes." For legibility, we consider a medium consisting of a single isotope where the atoms have no hyperfine structure, an axial magnetic field, and an electric field with two circular polarizations. The results of a general treatment, including effects neglected here, are given in Appendix C. As in the previous work, pressure effects and spontaneous emission from the upper to lower levels are neglected. The general state of loss anisotropy given in Ref. 1 is extended to include loss due to backscattering of one traveling-wave mode into the traveling wave of the opposite direction. Frequency- and amplitude-determining equations are given for both bidirectional and unidirectional Zeeman ring lasers.

The calculation is similar to that in the Zeeman theory,[1] with the exception of the algebraic method. We introduce the "perturbation tree," a graphical representation of the third-order components of the population matrix. This technique greatly simplifies the calculation and allows repetitious algebra to be abstracted in tables. The perturbation tree is given in general form and may be applied to any laser problem utilizing perturbation theory. The coefficients appearing in amplitude- and frequency-determining equations calculated via the perturbation tree are evaluated exactly and are given in a form which is computer oriented.

To begin our discussion, we give Maxwell's equations in a rotating frame to first order in $|v/c|$, give the form of the electric field and induced polarization, and obtain the self-consistency equations in Sec. II. The equation of motion of the population matrix and the expression for the polarization of the medium are derived in Sec. III. In Sec. IV the equation of motion for the population matrix is formally integrated to third order in the electric field. The perturbation tree is also introduced. First- and third-order contributions to the induced polarization are obtained Secs. V and VI, respectively. The amplitude- and frequency-determining equations are given in Sec. VII for the bidirectional case and in Sec. VIII for the unidirectional case.

II. ELECTROMAGNETIC FIELD EQUATIONS

In order to treat the ring laser including rotations, we must obtain Maxwell's equations for a reference frame in uniform rotation (see Fig. 1). Although the equations may be derived via the

general theory of relativity,[3-5] we are only concerned with perimeter speeds v much less than the speed of light. Hence we derive Maxwell's equations to first order in $|v/c|$. Following a treatment similar to Menegozzi and Lamb,[4] we assume that the observer in the rotating frame may relate his field quantities $\vec{E}(\vec{r}, t)$ and $\vec{B}(\vec{r}, t)$ to the fields $\vec{E}^0(\vec{r}^0, t^0)$ and $\vec{B}^0(\vec{r}^0, t^0)$ in the inertial frame by the equations of special relativity

$$\vec{E}^0(\vec{r}^0, t^0) = [\vec{E}(\vec{r}, t) - \vec{v}\times\vec{B}(\vec{r}, t)],$$
$$\vec{B}^0(\vec{r}^0, t^0) = [\vec{B}(\vec{r}, t) + \mu_0\epsilon_0\vec{v}\times\vec{E}(\vec{r}, t)], \tag{1}$$

where the physical measurements to determine $\vec{E}$ and $\vec{B}$ in the rotating frame are the same as those used to determine $\vec{E}^0$ and $\vec{B}^0$ in an inertial frame with any effects of inertial forces subtracted out.[4] The details of the transformation are given in Ref. 3. The macroscopic Maxwell equations for a rotating frame to first order in $|v/c|$ are found to be

$$\operatorname{div}(\vec{B} - \mu_0\epsilon_0\vec{v}\times\vec{E}) = 0,$$
$$\operatorname{curl}\vec{E} + \frac{\partial}{\partial t}[\vec{B} + \mu_0\epsilon_0(\vec{v}\times\vec{E})] = 0,$$
$$\operatorname{div}(\vec{E} - \vec{v}\times\vec{B}) = 0,$$
$$\operatorname{curl}\vec{B} - \mu_0\epsilon_0\left(\frac{\partial}{\partial t}\right)(\vec{E} - \vec{v}\times\vec{B}) = \mu_0\vec{J}, \tag{2}$$

where $\vec{v} = \vec{\Omega}_r\times\vec{r}$ is the instantaneous velocity at point $\vec{r}$ on the laser axis and $\vec{J}$ is the current density. With the usual replacement for nonmagnetic materials,[6]

$$\vec{J} \to \vec{J} + \frac{\partial\vec{P}}{\partial t}, \tag{3}$$

and Eq. (2), the wave equation to first order in $|v/c|$ becomes

$$\operatorname{curl}\operatorname{curl}\vec{E} + \mu_0\epsilon_0\left(\frac{\partial^2\vec{E}}{\partial t^2}\right) + \mu_0\epsilon_0\left(\frac{\partial}{\partial t}\right)[\operatorname{curl}(\vec{v}\times\vec{E}) + \vec{v}\times\operatorname{curl}\vec{E}] = -\mu_0\left(\frac{\partial}{\partial t}\right)\left(\vec{J} + \frac{\partial\vec{P}}{\partial t}\right), \tag{4}$$

where the induced polarization of the active medium $\vec{P}$ is described in the rest frame of the laser cavity. Following the treatment of Sargent *et al.*,[1] we choose a current density to provide for different cavity resonance frequencies and losses for linearly polarized radiation along unit vectors $\hat{i}$ and $\hat{j}$, circular birefringence, and different effective cavity lengths for different polarizations and traveling waves. Since we are concerned with a traveling-wave laser, the possibility exists for backscattering of one traveling-wave mode into a mode of the opposite direction. This may be accounted for by including a loss term $\bar{\sigma}_s\cdot\vec{E}$ in the current density. These considerations lead to

$$\vec{J} = \left[\bar{\sigma}_s + \bar{\sigma}' - \nu^{-1}\bar{\sigma}''\left(\frac{\partial}{\partial t}\right)\right]\cdot\vec{E}, \tag{5}$$

where $\bar{\sigma}'$ and $\bar{\sigma}''$ are conductivity tensors.

Neglecting the transverse spatial variations in $\vec{E}$, we find $\operatorname{curl} \to \hat{s}\,\partial/\partial s$, which gives

$$\operatorname{curl}\operatorname{curl}\vec{E} \simeq -\frac{\partial^2\vec{E}}{\partial s^2}. \tag{6}$$

With these simplifications and Eq. (5), the wave equation (4) reduces to

$$-\frac{\partial^2\vec{E}}{\partial s^2} + \mu_0\left[\bar{\sigma}_s + \bar{\sigma}' - \nu^{-1}\bar{\sigma}''\left(\frac{\partial}{\partial t}\right)\right]\cdot\frac{\partial\vec{E}}{\partial t} - 2\mu_0\epsilon_0[(\vec{\Omega}_r\times\vec{r})\cdot\hat{s}]\frac{\partial^2\vec{E}}{\partial s\,\partial t} = -\mu_0\frac{\partial^2\vec{P}}{\partial t^2}, \tag{7}$$

where $\hat{s}$ is the coordinate along the laser axis (as shown in Fig. 1).

The electric field may be expanded in the form

$$\vec{E}(s, t) = \frac{1}{2}\sum_{i=1}^{2}\sum_{n}(\hat{e}_i E_{ni}(t) \times \exp\{-i[\nu_{ni}t + \phi_{ni}(t)]\}U_{ni}(s) + (\text{c.c.})), \tag{8}$$

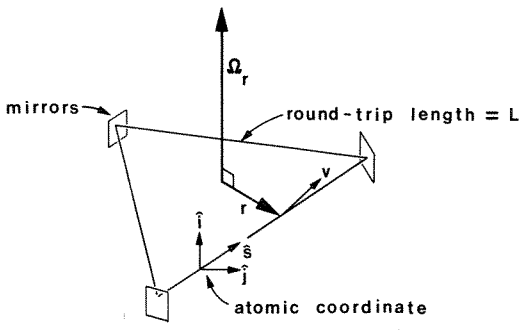

FIG. 1. Possible ring-laser geometry and the position of the vectors $\vec{\Omega}_r$, $\vec{v}$, $\vec{r}$, and $\hat{s}$ used in the derivation of Eqs. (2), (4), and (7). Here $\vec{\Omega}_r$ is the rotation vector, $\vec{v}$ is the instantaneous velocity at point $\vec{r}$ on the laser axis and is equal to $\vec{\Omega}_r\times\vec{r}$ and $\hat{s}$ is the unit vector along the laser axis. The unit vectors $\hat{i}$ and $\hat{j}$ are used to describe the polarization of the electric field. In the derivation of the electromagnetic field equations it is important to note that we require the vector $\vec{\Omega}_r$ to be perpendicular to the plane of the ring laser.

where $\hat{e}_i$ are any two orthogonal unit vectors in the transverse plane and may be complex. The amplitudes $E_{ni}(t)$ and phases $\phi_{ni}(t)$ are real functions which vary little in an optical period. In this treatment we allow the subscript n to index *both* Fox-Li quasimodes and traveling waves. Hence a "mode" refers to a single Fox-Li quasimode traveling in a single direction (clockwise or counterclockwise). Then the $U_{ni}(s)$ are the complex eigenfunctions corresponding to the nth mode for e_i. We use $U_{ni}(s) = e^{id_n K_{ni} s}$ in this treatment, where $d_n = +1(-1)$ for the clockwise (counterclockwise) traveling wave. Here we have not explicitly included any phase shifts at the mirrors, and we restrict our discussion to cavities with an active medium in only one segment of the ring. Polarization anisotropic cavities such as the three-mirror configuration may be made isotropic by the insertion of a half-wave plate in a vacant segment to compensate for the π phase shift of the polarization in the plane of the ring at the mirrors.[7] More complicated phase shifts may be accounted for in the general anisotropy matrix $\vec{G}$ discussed shortly, or by the inclusion of additional phenomenological terms in the current density (5). The polarization may similarly be written in the form

$$\vec{P}(s,t) = \frac{1}{2}\sum_{i=1}^{2}\sum_{n}\{\hat{e}_i \mathcal{P}_{ni}(t)\exp\{-i[\nu_{ni}t + \phi_{ni}(t)]\} \times U_{ni}(s) + (\text{c.c.})\}, \quad (9)$$

where $\mathcal{P}_{ni}(s,t)$ is a slowly varying complex function of time. $\mathcal{P}_{ni}(t)$ is referred to as the complex polarization.

In obtaining the self-consistency equations, we drop the complex conjugates of the expressions for $\vec{E}$ and $\vec{P}$, which amounts to making the rotating-wave approximation in the derivation of $\vec{P}$ and equating the coefficients of the positive frequency term $e^{-i\nu t}$ to zero separately from those of the negative frequency term $e^{i\nu t}$. Then one substitutes Eqs. (8) and (9) without complex conjugates into Eq. (7), projects the result onto the unit vectors $\hat{e}_i$, and then onto the mode $U_{ni}(s)$. In doing so, we neglect rapidly varying terms containing $\ddot{E}_{ni}$, $\ddot{\phi}_{ni}$, $\sigma_{ii'}\dot{\phi}_{ni'}$, $\sigma_{sii'}\dot{\phi}_{ni'}$, $\sigma_{ii'}\dot{E}_{ni'}$, and $\dot{\phi}_{ni}\dot{E}_{ni}$.

Carrying out the indicated operations, noting that

$$(\nu_{ni} + \dot{\phi}_{ni})^2 - \Omega_n \sim 2\nu(\nu_{ni} + \dot{\phi}_{ni} - \Omega_n),$$

and dividing through by

$$-(2\nu)\exp[-i(\nu_{ni}t + \phi_{ni})],$$

we find the component equation

$$(\nu_{ni} + \dot{\phi}_{ni} - \Omega_n)E_{ni} + iE_{ni} + L^{-1}d_n K_{ni}\int_0^L ds\,(\vec{\Omega}_r \times \vec{r})\cdot\hat{s} + i\frac{1}{2}\nu\sum_{\mu}\sum_{i'}\hat{e}_i\cdot\left(L^{-1}\int_0^L ds\,U_{ni}^*(s)\,(\vec{\sigma} + \vec{\sigma}_s)U_{ni'}(s)\right)\cdot\hat{e}_{i'}E_{\mu i'}e^{i\psi_{ni,\mu i'}} = \frac{1}{2}\left(\frac{\nu}{\epsilon_0}\right)\mathcal{P}_{ni}, \quad (10)$$

where L is the round-trip cavity length, $\psi_{ni,\mu i'} = (\nu_{ni} - \nu_{\mu i'})t + \phi_{ni} - \phi_{\mu i'}$ are relative phase angles, and $\vec{\sigma} = \vec{\sigma}' + i\vec{\sigma}''$.

The vector identity $(\vec{A}\times\vec{B})\cdot\vec{C} = \vec{A}\cdot(\vec{B}\times\vec{C})$ reduces the first integral in Eq. (10) to

$$\int_0^L ds\,(\vec{\Omega}_r\times\vec{r})\cdot\hat{s} = \vec{\Omega}_r\cdot\int_0^L(\vec{r}\times d\vec{s}) = 2(\vec{\Omega}_r\cdot\vec{A}) = 2\Omega_r A, \quad (11)$$

where $\vec{A}$ is a vector with magnitude equal to the area of the polygon formed by the laser and direction normal to the plane of this polygon (parallel to $\vec{\Omega}_r$). For the second integral it is convenient to define an "anisotropy matrix" which includes backscattering effects:

$$\vec{G} = (\epsilon_0\nu L)^{-1}\int_0^L ds\,U_{ni}^*(s)\,(\vec{\sigma} + \vec{\sigma}_s)U_{\mu i'}(s). \quad (12)$$

If backscattering is not considered, and $\vec{\sigma}$ is a constant, $n = \mu$ and this anisotropy matrix reduces to the form used by Sargent *et al.*[1] In the present context, the real parts of the diagonal terms are the reciprocals of cavity Q's, and the imaginary parts are frequency displacements of the ith polarization from the nonbirefringent cavity frequency $n\pi c/L$. The off-diagonal terms couple the modes via backscattering and polarization anisotropies. The summation over μ only includes terms for which n and μ index the same Fox-Li quasimode, for the other terms lead to rapidly varying phase angles which the electric field amplitudes cannot follow (high-Q cavity). If the frequencies of the traveling-wave modes (centered about a Fox-Li quasimode) are significantly different, several terms may be dropped due to rapidly varying phase angles. This corresponds to an inability of backscattered radiation to contribute to a mode if the frequencies of the mode and backscattered radiation are not approximately equal (see Ref. 8). Recall that subscripts index Fox-Li quasimodes, traveling-wave modes, and

polarizations. The component equation [Eq. (10)] may then be written

$$(\nu_n+\dot{\phi}_n-\Omega_n)E_n+i\left(E_n+\frac{1}{2}\nu\sum_\mu g_{n\mu}e^{i\psi_{n\mu}}E_\mu\right)=\frac{1}{2}\left(\frac{\nu}{\epsilon_0}\right)\mathcal{P}_n\,, \quad (13)$$

where

$$\Omega_n=\Omega_n-2d_nK_n\Omega_r(A/L)\,,$$
$$\psi_n=(\nu_n-\nu_\mu)t+\phi_n-\phi_\mu\,. \quad (14)$$

Equating the real and imaginary parts separately to zero, we arrive at the self-consistency equations

$$\dot{E}_n+\frac{1}{2}\nu\sum_\mu \mathrm{Im}(ig_ne^{i\psi_{n\mu}})E_\mu=-\left(\frac{\nu}{\epsilon_0}\right)\mathrm{Im}(\mathcal{P}_n)\,, \quad (15a)$$

$$\nu_n+\dot{\phi}_n=\Omega_n-\frac{1}{2}\nu\sum_\mu \mathrm{Re}(ig_{n\mu}e^{i\psi_{n\mu}})\times E_\mu E_n^{-1}-\frac{1}{2}\left(\frac{\nu}{\epsilon_0}\right)E_n^{-1}\mathrm{Re}(\mathcal{P}_n)\,. \quad (15b)$$

If one neglects backscattering and the resulting $\bar{G}$ matrix is diagonal in the representation specified by $\hat{e}_1$ and $\hat{e}_2$, these equations reduce to

$$\dot{E}_n+\frac{1}{2}\left(\frac{\nu}{Q_n}\right)E_n=-\frac{1}{2}\left(\frac{\nu}{\epsilon_0}\right)\mathrm{Im}(\mathcal{P}_n)\,, \quad (16a)$$

$$\nu_n+\dot{\phi}_n=\Omega_{n'}-\frac{1}{2}\left(\frac{\nu}{\epsilon_0}\right)E_n^{-1}\mathrm{Re}(\mathcal{P}_n)\,, \quad (16b)$$

where

$$\Omega_{n'}=\Omega_n+\tfrac{1}{2}\nu\,\mathrm{Im}(g_{nn})\,. \quad (17)$$

If the losses are independent of polarization and traveling wave, Eqs. (16) describe the amplitudes and frequencies for any set of orthogonal unit vectors.

Two useful bases for formulating the problem are the x-y basis for which we have

$$\hat{e}_1=\hat{\imath}\,,\quad \hat{e}_2=\hat{\jmath}\,, \quad (18)$$

and the ± (circularly polarized) basis for which we have

$$\hat{e}_1=\hat{\epsilon}_-=2^{-1/2}(\hat{\imath}+i\hat{\jmath})\,,$$
$$\hat{e}_2=\hat{\epsilon}_+=2^{-1/2}(\hat{\imath}-i\hat{\jmath})\,. \quad (19)$$

Since we are concerned mainly with axial magnetic field, we use the ± basis for the remainder of the calculation. As explained in Ref. 1, this is especially convenient for electric-dipole transitions in which the magnetic-quantum-number changes by +1(−1) contribute to $E_{n+}(E_{n-})$. Properties of transverse magnetic fields and transformations of the $\bar{G}$ matrix are discussed in detail in Ref. 1.

III. POLARIZATION OF ATOMIC MEDIUM

The laser action takes place between two atomic levels a and b, as depicted in Fig. 2. The levels are connected by an electric-dipole transition and may be characterized by the total angular momentum J_a and J_b and other quantum numbers n_a and n_b, respectively. The inclusion of isotopes with nuclear spin and hyperfine structure is straightforward but algebraically messy. Since they were included by Sargent *et al.*,[1] they are not treated here. However, the completely general expressions in Appendix C contain both considerations.

A basis for a matrix representation consists of eigenvectors

$$|nJm\rangle\,, \quad (20)$$

where $n=n_a,\ n_b$, $J=J_a,\ J_b$, and m runs over the corresponding magnetic sublevels of a and b. We assume the decay operator Γ is diagonal in this basis with elements $\Gamma_{mm}=\Gamma_m$. The Hamiltonian $\mathcal{H}$ has diagonal elements

$$\hbar\omega_{a'}=\hbar\omega_a+\mu_BHg_aa'\,,$$
$$\hbar\omega_{b'}=\hbar\omega_b+\mu_BHg_bb'\,, \quad (21)$$

where a' and b' are magnetic quantum numbers of the levels a and b, respectively, $\hbar\omega_a$ and $\hbar\omega_b$ are the zero-field energies of a and b, g_a and g_b are the Landé g factors for the levels, H is the magnetic field strength, and μ_B is the Bohr magneton. The off-diagonal elements of the Hamil-

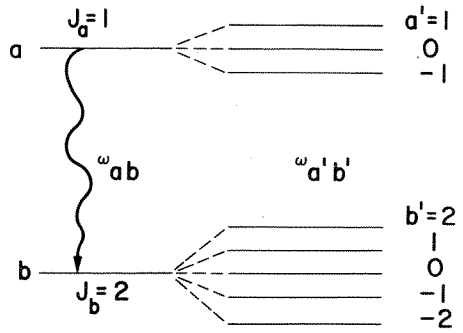

FIG. 2. Possible level scheme showing how levels a and b might be split into $2J_a+1=3$ and $2J_b+1=5$ sublevels, respectively, by an applied dc magnetic field. Here J_a and J_b are the total angular momenta of the levels a and b; a' and b' are the corresponding magnetic quantum numbers; ω_{ab} is the zero field optical frequency between levels a and b, and $\omega_{a'b'}$ is a representative optical frequency in the presence of a magnetic field. This level diagram corresponds to the 1.15-, 3.39-, and 0.6328-μ Ne line in the He-Ne laser.

tonian $V_{a'b'}$ are the matrix elements of the time-dependent perturbation energy associated with the electric field

$$V_{a'b'} = -\langle n_a J_a a' | e\vec{\mathbf{E}} \cdot \vec{\mathbf{r}} | n_b J_b b' \rangle , \tag{22}$$

where e is the charge of the electron and $\vec{\mathbf{r}}$ is the position vector of the atom

$$\vec{\mathbf{r}} = x\hat{i} + y\hat{j} + s\hat{\mathbf{s}} . \tag{23}$$

The position vector $\vec{\mathbf{r}}$ may be written in spherical coordinates with complex combinations of $\hat{i}$ and $\hat{j}$ which make the calculation of matrix elements particularly easy:

$$\vec{\mathbf{r}} = \tfrac{1}{2}\hat{r}\sin\phi[(\hat{i} - i\hat{j})e^{i\phi} + (\hat{i} + i\hat{j})e^{-i\phi}] + r\cos\phi\hat{\mathbf{s}} , \tag{24}$$

where θ and ϕ are the standard polar and azimuthal angles of $\hat{r}$ with respect to the atomic axis $\{\hat{i}, \hat{j}, \hat{\mathbf{s}}\}$. Writing Eq. (24) in terms of $\hat{\epsilon}_-$ and $\hat{\epsilon}_+$ given by Eq. (19) and substituting the result into Eq. (23), one has

$$V_{a'b'} = -2^{1/2}\wp_{a'b}(\vec{\mathbf{E}} \cdot \hat{\epsilon}_+ \delta_{a'b'+1} + \vec{\mathbf{E}} \cdot \hat{\epsilon}_- \times \delta_{a',b'-1} + \vec{\mathbf{E}} \cdot \hat{\mathbf{s}}\delta_{a'b'}) , \tag{25}$$

where the dipole matrix elements $\wp_{a'b'}$ are given by

$$\begin{aligned}
\wp_{a'b'} &= \langle J_a a' | \tfrac{1}{2} e r \sin\theta e^{\pm i\phi} | J_b b' \rangle \\
&\quad (a' = b' \pm 1, \quad J_a = J_b \pm 1, 0) \\
&= \langle J_a a' | e r \cos\theta | J_b b' \rangle \\
&\quad (a' = b', \quad J_a = J_b \pm 1, 0) \\
&= 0 \\
&\quad \text{otherwise.}
\end{aligned} \tag{26}$$

With explicit values (Condon and Shortley[9]), we have

$$\begin{aligned}
\wp_{a'b'} &= \mp\tfrac{1}{2}\wp[(J_b \pm a')(J_b \pm a' + 1)]^{1/2} \\
&\quad (a' = b' \pm 1, \quad J_a = J_b - 1) \\
&= \wp[J_b^2 - a'^2]^{1/2} \\
&\quad (a' = b', \quad J_a = J_b - 1) \\
&= \tfrac{1}{2}\wp[(J_a \mp a')(J_a \pm a' + 1)]^{1/2} \\
&\quad (a' = b' \pm 1, \quad J_a = J_b) \\
&= \wp a' \\
&\quad (a' = b', \quad J_a = J_b) \\
&= \mp\tfrac{1}{2}\wp[(J_a \pm b')(J_a \pm b' + 1)]^{1/2} \\
&\quad (a' = b' \pm 1, \quad J_a = J_b + 1) \\
&= \wp[J_a^2 - b'^2]^{1/2} \\
&\quad (a' = b', \quad J_a = J_b + 1) ,
\end{aligned} \tag{27}$$

where $\wp$ is the reduced matrix element $(n_a J_a \| er \| n_b J_b)$. Substituting Eq. (8), expressed in the $\hat{\epsilon}_\pm$ basis, into Eq. (25), one has

$$\begin{aligned}
V_{a'b'} &= -2^{-1/2}\wp_{a'b'} \sum_\mu \delta_{a',b'+p_\mu} \\
&\quad \times E_\mu \exp[-i(\nu_\mu t + \phi_\mu)]U_\mu(s) + (\text{c.c.}) ,
\end{aligned}$$

$$V_{b'a'} = V^*_{a'b'} , \tag{28}$$

where the polarizations $p_\mu = +1(-1)$ for right (left) circularly polarized light.

The equation of motion of a density matrix $\rho(\alpha, s_0, t_0, v)$ describing the state in which a single atom is excited to the eigenstate α (i.e., $|nJm\rangle$), at place s_0, time t_0, and s component of velocity v, is

$$\dot{\rho}(\alpha, s_0, t_0, v, t) = -(i/\hbar)[\mathcal{H}, \rho] - \tfrac{1}{2}[\Gamma\rho + \rho\Gamma] , \tag{29}$$

where $\mathcal{H}$ is the Hamiltonian and Γ is the decay operator mentioned earlier. With this density operator, the average electric-dipole moment $\vec{\mathbf{p}}$ is

$$\vec{\mathbf{p}} = \mathrm{Tr}(\rho e\vec{\mathbf{r}}) . \tag{30}$$

All atoms arriving at s at time t, regardless of s_0, t_0, α, and v contribute to the macroscopic polarization $\vec{\mathbf{P}}(s, t)$ as follows[1]:

$$\vec{\mathbf{P}}(s, t) = \sum_\alpha \int_{-\infty}^{t} dt_0 \int_0^L ds_0 \int_0^\infty dv\, \lambda_\alpha(s_0, v, t_0) \times \mathrm{Tr}(\rho e\vec{\mathbf{r}})\delta[s - s_0 - v(t - t_0)] , \tag{31}$$

where we have assumed that the excitation mechanism always excites an atom to the eigenstate α, and $\lambda_\alpha(s_0, v, t_0)$ is the number of atoms excited to the eigenstate α per unit time per unit volume.

The interaction energy (25) does not depend[2] on s_0, t_0, or α but only on s, t, and v. Proceeding as in Ref. 1, we use this fact to average the pure case over s_0, t_0, and α before the integration of the equations of motion. Specifically, we define the population matrix

$$\rho(s, v, t) = \sum_\alpha \int_{-\infty}^{t} dt_0 \int_0^L ds_0\, \lambda_\alpha(s_0, v, t_0) \times \rho(\alpha, s_0, t_0, v, t)\delta[s - s_0 - v(t - t_0)] , \tag{32}$$

where s, t, and v label an ensemble of atoms with velocity v which at time t arrive at place s.

Differentiating Eq. (32) with respect to t we find the equation of motion for $\rho(s, v, t)$. Using

the Leibnitz rule one has

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)\rho=\lambda(s,v,t)+\sum_{\alpha}\int_{-\infty}^{t}dt_0\int_0^L ds_0\,\lambda_\alpha(s,v,t)\times\dot{\rho}(\alpha,s_0,t_0,v,t)\delta[z-z_0-v(t-t_0)]\,. \quad (33)$$

Here we have assumed that the (diagonal) excitation operator $\lambda(s,v,t)$, with elements $\lambda_\alpha(s_0,v,t_0)$ which are slowly varying in time and space, may be evaluated at s, t, v. Using Eq. (29) for $\dot{\rho}(\alpha, s_0, t_0, v, t)$ in Eq. (33), interchanging the order of the Hamiltonian operator and the commutation operation with the integration, and invoking the definition of the population matrix, Eq. (32), we find the equation of motion for the population matrix,

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)\rho(s,v,t)=-(i/\hbar)[\mathcal{H},\rho(s,v,t)]-\frac{1}{2}[\Gamma\rho(s,v,t)+\rho\Gamma]+\lambda(s,v,t)\,. \quad (34)$$

Combining the definition of $\rho(s,v,t)$ [Eq. (32)] with Eq. (31), we find that $\vec{\mathrm{P}}(s,t)$ becomes

$$\vec{\mathrm{P}}(s,t)=\int_{-\infty}^{\infty}dv\,\mathrm{Tr}[\rho(s,v,t)e\vec{\mathbf{r}}]\,. \quad (35)$$

We obtain the equation for the complex polarization $\mathcal{P}_n(t)$ by equating the positive frequency parts of Eqs. (9) and (35), projecting onto $U_n(s)$, and multiplying through by $2\exp[-i(\nu_n t+\phi_n)]$. We find

$$\mathcal{P}_n(t)=2\exp[-i(\nu_n t+\phi_n)]L^{-1}\int_0^L ds\,U_n^*(s)\times\int_{-\infty}^{\infty}dv\,\mathrm{Tr}\{\rho(s,v,t)e[\hat{e}(p_n)\cdot\vec{\mathbf{r}}]\}\,. \quad (36)$$

Since $\hat{e}(p_n)\cdot\vec{\mathbf{r}}$ yields a matrix element given by $2^{-1/2}$ times the complex conjugate of Eq. (27), Eq. (36) reduces to

$$\mathcal{P}_n(t)=2\sqrt{2}\sum_{a'}\sum_{b'}\exp[-i(\nu_n t+\phi_n)]\wp_{b'a'}\delta_{a',b'+p_n}\times L^{-1}\int_0^L ds\,U_n^*(s)\int_0^{\infty}dv\,\rho_{a'b'}(s,v,t)\,. \quad (37)$$

IV. INTEGRATION OF EQUATIONS OF MOTION

Inserting the total Hamiltonian [Eqs. (21) and (22)] into the equation of motion (34), one obtains the components

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)\rho_{a'b'}=-(i\omega_{a'b'}+\gamma_{a'b'})\rho_{a'b'}+\frac{i}{\hbar}\left(\sum_{a''}V_{a''b'}\rho_{a'a''}-\sum_{b''}V_{a'b''}\rho_{b''b'}\right), \quad (38)$$

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)\rho_{a'a''}=-(i\omega_{a'a''}+\gamma_{a'b''})\rho_{a'a''}+\left(\frac{i}{\hbar}\right)\sum_{b''}(V_{b''a''}\rho_{a'b''}-V_{a'b''}\rho_{b''a''})+\lambda_{a'}\delta_{a'a''}\,, \quad (39)$$

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)\rho_{b''b'}=-(i\omega_{b''b'}+\gamma_{b''b'})\rho_{b''b'}+\left(\frac{i}{\hbar}\right)\sum_{a''}(V_{a''b'}\rho_{b''a''}-V_{b''a''}\rho_{a''b'})+\lambda_{b'}\delta_{b'b''}\,, \quad (40)$$

$$\rho_{b'a'}=\rho_{a'b'}^*\,, \quad (41)$$

where a'' and b'' are magnetic quantum numbers for the sublevels of a and b, respectively,

$$\omega_{\alpha\alpha'}=\omega_\alpha-\omega_{\alpha'}\,,\qquad \gamma_{\alpha\alpha'}=\tfrac{1}{2}(\gamma_\alpha+\gamma_{\alpha'})+\gamma_{\text{phase}}\,,\qquad \alpha,\alpha'=a',b',a'',b''\,, \quad (42)$$

where γ_{phase} is the reciprocal of the phase diffusion time T_2. The iteration procedure to obtain Eqs. (38)–(41) to any desired order in the perturbation $V_{a'b'}$ formally proceeds along the lines of Refs. 1 and 2. Equations (38)–(40) have the form

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)f(s,v,t)=g(s,v,t)\,.$$

As can be verified by direct substitution, this equation has the formal integral

$$f(s,v,t)=\int_{-\infty}^{t}dt'\,g(s',v,t')\,,$$

where

$$s'=s-v(t-t')\,.$$

Assuming the excitation rates have a separable form

$$\lambda_\alpha(s,v,t)=W(v)\Lambda_\alpha(s,t)\,, \quad (43)$$

and defining the excitation rate density

$$N_{a'b'}(s,t)=\Lambda_{a'}(s,t)\gamma_{a'}^{-1}-\Lambda_{b'}(s,t)\gamma_{b'}^{-1}\,, \quad (44)$$

where $\Lambda_{a'}(s,t)$ and $\Lambda_{b'}(s,t)$ are the unsaturated population inversion densities for magnetic sublevels a' and b', respectively, we find the first-order contribution to $\rho_{a'b'}$:

$$\rho_{a'b'}^{(1)}(s,v,t)=(i/\hbar)W(v)N_{a'b'}(s,t)\times\int_0^{\infty}d\tau'\,V_{a'b'}(s',t')\exp[-(\gamma_{a'b'}+i\omega_{a'b'})\tau']\,. \quad (45)$$

Here $W(v)$ is the velocity distribution of the atoms, $t'=t-\tau'$, and $s'=s-v\tau'$. Substitution of Eq. (45)

into Eqs. (39) and (40) yields the second-order components of $\rho_{a'a''}$ and $\rho_{b''b'}$ (see Ref. 1 for values). In turn, substituting the second-order results for $\rho_{a'a''}$ and $\rho_{b''b'}$ in Eq. (35), one has the third-order components

$$\rho^{(3)}_{a'b'}(s,v,t) = -\left(\frac{i}{\hbar^3}\right) W(v) \int_0^\infty d\tau' \int_0^\infty d\tau'' \int_0^\infty d\tau''' \exp[-(\gamma_{a'b'}+i\omega_{a'b'})\tau']$$

$$\times \sum_{a''}\sum_{b''} (V_{a''b'}(s',t') \exp[-(\gamma_{a'a''}+i\omega_{a'a''})\tau']\{V_{b''a''}(s'',t'')N_{a'b''} \exp[-(\gamma_{a'b''}+i\omega_{a'b''})\tau''']V_{a'b''}(s''',t''')$$

$$- V_{a'b''}(s'',t'')N_{b''a''} \exp[-(\gamma_{b''a''}+i\omega_{b''a''})\tau''']V_{b''a''}(s''',t''')\} - V_{a'b''}(s',t') \exp[-(\gamma_{b''b'}+i\omega_{b''b'})\tau'']$$

$$\times\{V_{a''b'}(s'',t'')N_{b''a''} \exp[-(\gamma_{b''a''}+i\omega_{b''a''})]V_{b''a''}(s''',t''')$$

$$- V_{b''a''}(s'',t'')N_{a''b'} \exp[-(\gamma_{a''b'}+i\omega_{a''b'})\tau''']V_{a'b'}(s''',t''')\}), \qquad (46)$$

where

$$\begin{aligned} t'' &= t' - \tau'' = t - \tau' - \tau'', \\ s'' &= s' - v\tau' = s - v(\tau' + \tau''); \\ t''' &= t'' - \tau''' = t - \tau' - \tau'' - \tau''', \\ s''' &= s'' - v\tau''' = s - v(\tau' + \tau'' + \tau'''). \end{aligned} \qquad (47)$$

Equation (46) may conveniently be represented by a perturbation tree, as shown in Fig. 3, where we have defined

$$\begin{aligned} e_{\alpha\alpha'} &= \exp[-(\gamma_{\alpha\alpha'}+i\omega_{\alpha\alpha'})\tau'], \\ \alpha, \alpha' &= a', b', a'', b''. \end{aligned} \qquad (48)$$

In our tree connecting lines and vertical ascent indicate multiplication, and horizontal rows indicate addition. The earliest perturbation, contributing to $\rho^{(0)}$ at time t''', connecting states $|n_aJ_aa'\rangle$ and $|n_bJ_bb''\rangle$, $|n_bJ_bb''\rangle$ and $|n_bJ_ba''\rangle$, $|n_aJ_aa''\rangle$ and $|n_bJ_bb'\rangle$, is represented by the first (bottom) row of boxes. The first-order contribution connecting $|n_bJ_bb''\rangle$ and $|n_aJ_aa'\rangle$, $|n_aJ_aa'\rangle$ and $|n_bJ_bb''\rangle$, $|n_aJ_aa''\rangle$ and $|n_bJ_bb'\rangle$, at time t'' is represented by the second row. Finally, the third-order contribution connecting $|n_aJ_aa''\rangle$ and $|n_bJ_bb'\rangle$, $|n_aJ_aa'\rangle$ and $|n_bJ_bb''\rangle$, is represented by the third row. The $e_{\alpha\alpha'}$ are the integrating factors which arise in the formal integration of Eqs. (38)–(41). The subscripts μ, ρ, and σ are used for the third-order contribution to the complex polarization $\mathcal{P}_n$, and the $t=1$ which appear at the bottom are subscripts to integrals which appear in $\mathcal{P}_n$ (see Sec. VI). Up to this point, we have not explicitly included the form of $V_{a'b'}$. Thus our perturbation tree is per-

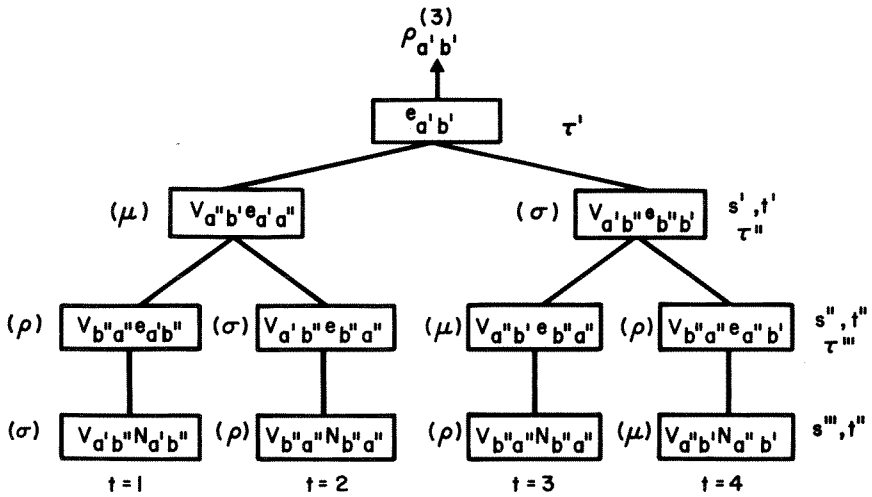

FIG. 3. Perturbation tree described in Sec. IV is shown. The $e_{\alpha\alpha'}$ are defined by Eq. (48). In the tree, connecting lines and vertical ascent indicate multiplication and horizontal rows indicate addition. The earliest perturbation is represented by the first (bottom) row of boxes. The first-order contribution is represented by the second row and finally, the third-order contribution is given by the third row. The $e_{\alpha\alpha'}$ are integrating factors which arise from the formal integration of the equations of motion for the population matrix. Since we have left the perturbation $V_{a'b'}$ in a general form, the tree may be used in any laser problem employing third-order perturbation theory by using the correct $V_{a'b'}$. Higher-order perturbation theory may be used if the tree is expanded appropriately.

fectly general for any laser calculation involving third-order perturbation theory.[2] In particular, by specifying the correct $U_n(s)$ one may do calculations for (i) unidirectional ring laser by setting $U(s)=e^{iks}$ with only one traveling wave, (ii) standing-wave two-mirror laser by setting $U(z)=\sin(Kz)$, where z is along the two-mirror-laser axis, and (iii) the bidirectional ring laser by setting $U(s)=e^{\pm iks}$ and considering oppositely directed traveling waves. We do (iii), the most general case, and specialize to (i) and (ii) as a check on our calculations. The standing-wave two-mirror laser was treated by Sargent *et al.*[1] Although the unidirectional ring laser may be obtained by simplifying the standing-wave analysis, to our knowledge this has not been done and is presented here.

V. FIRST-ORDER THEORY

The first-order calculation formally proceeds in exactly the same manner as the standing-wave two-mirror theory presented in Ref. 1, with the exception of the $U_n(s)$ and the convention that subscripts index modes as defined earlier. We shall assume a Maxwellian velocity distribution described by

$$W(v)=[(\pi)^{1/2}u]^{-1}e^{-(v/u)^2}, \tag{49}$$

where u is the most probable atomic speed. Combining Eqs. (28), (45), and (37), and using the explicit form for $U_n(s)$, one has

$$\mathcal{P}_n^{(1)}(t)=-2\left(\frac{i}{\hbar}\right)\sum_{a'}\sum_{b'}|\wp_{a'b'}|^2\sum_{\mu}\delta_{a',b'+p_n}\delta_{a',b'+p_\mu}$$
$$\times L^{-1}\int_0^L ds N_{a'b'}(s,t)\exp[-i(d_nK_n-d_\mu K_\mu)s]\exp\{i[(\nu_n-\nu_\mu)t+\phi_n-\phi_\mu]\}\int_{-\infty}^{\infty}dv\,W(v)\int_0^{\infty}d\tau'$$
$$\times\exp\{-i[\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu)+id_\mu K_\mu v]\tau'\}+(\text{a term containing }\exp\{i[(\nu_n+\nu_\mu)t+\phi_n+\phi_\mu]\}), \tag{50}$$

where we have freely interchanged the order of summation and integration. In order that the integrand of the integral over s vary little in an optical period we take $d_n=d_\mu$. Since the velocity distribution is an even function in v, the d_μ do not affect the integral over v, and may be dropped. Although the Kronecker δ's $\delta_{a'b'+p_n}$, $\delta_{a',b'+p_n}$ require that $p_n=p_\mu$, we choose not to make this simplification at this point. Dropping the rapidly varying term and performing the time and velocity integrals, we find the first-order contribution to the complex polarization

$$\mathcal{P}_n^{(1)}(t)=-2(\hbar Ku)^{-1}\sum_{\mu}E_\mu e^{i\psi_{n\mu}}$$
$$\times\sum_{a'}\sum_{b'}\delta_{a',b'+p_n}\delta_{a',b'+p_\mu}\delta_{d_n,d_\mu}$$
$$\times|\wp_{a'b'}|^2N_{a'b',d_n(n-\mu)}Z[\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu)], \tag{51}$$

where the relative phase angle is

$$\psi_{n\mu}=(\nu_n-\nu_\mu)t+\phi_n-\phi_\mu, \tag{52}$$

the plasma dispersion function is

$$Z(v)=iK\pi^{-1/2}\int_{-\infty}^{\infty}dv\exp[-(v/u)^2]$$
$$\times(v\pm iKv)^{-1}, \tag{53}$$

and the Fourier components of the population inversion density are

$$N_{a'b',d_n(n-\mu)}(t)=L^{-1}\int_0^L ds N_{a'b'}(s,t)$$
$$\times\exp[-id_n(K_n-K_\mu)s]. \tag{54}$$

We will frequently use the abbreviation

$$N_{a'b'}\equiv N_{a'b',(0)}(t)$$
$$=L^{-1}\int_0^L ds N_{a'b'}(s,t). \tag{55}$$

Substitution of Eq. (51) into the self-consistency equations (15a) and (15b) gives

$$\dot{E}_n=\sum_{\mu}\mathrm{Im}(\bar{\alpha}_{n\mu}e^{i\psi_{n\mu}})E_\mu, \tag{56}$$

$$\nu_n+\dot{\phi}_n=\Omega_n+\sum_{\mu}\mathrm{Re}(\bar{\alpha}_{n\mu}e^{i\psi_n})E_\mu E_n^{-1}, \tag{57}$$

where

$$\bar{\alpha}_{n\mu}=-\frac{1}{2}i\nu g_{n\mu}+\nu(\hbar KuL\epsilon_0)^{-1}\sum_{a'}\sum_{b'}\delta_{a',b'+p_n}\delta_{a',b'+p_\mu}$$
$$\times\delta_{d_n,d_\mu}|\wp_{a'b'}|^2N_{a'b',d_n(n-\mu)}Z[\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu)]. \tag{58}$$

If the frequency difference between opposite traveling waves on the same Fox-Li quasimode is large enough to make terms for which $d_n\neq d_\mu$ contain rapidly varying phase angles, Eqs. (56) and (57) reduce to equations similar to those given by

Sargent *et al.* (Ref. 1), namely, the summations over $\mu \rightarrow i$.[1]

VI. THIRD-ORDER THEORY

To obtain the third-order contribution to the complex polarization $\mathcal{P}_n(t)$ it is necessary to substitute the explicit form of $V_{a'b'}$ into Eq. (46) and use that result in Eq. (37). The substitution of Eq. (28) for $V_{a'b'}$ may be greatly simplified if it is noted that only positive-frequency terms in $V_{a'b'}$ lead to resonant contributions to the density matrix elements. In the same manner, only negative-frequency terms of $V_{b'a'}$ lead to resonant contributions. The "perturbation tree" of Fig. 3 simplifies the algebra of the indicated operation considerably. We associate subscripts μ with $V_{a''b'}$, ρ with $V_{b''a''}$, and σ with $V_{a'b''}$. Although the times of perturbation for μ, ρ, and σ change from branch to branch in Fig. 3, the signs do not; μ and σ are associated with positive-frequency terms and ρ with negative-frequency terms. Each perturbation $V_{a'b'}$ consists of a right ($V^r_{a'b'}$) and left ($V^l_{a'b'}$) traveling wave, e.g., $V_{a'b'} = V^l_{a'b'} + V^r_{a'b'}$. Hence in the third order we have eight possible subbranches for each branch in Fig. 3. Because of the atomic motion, the perturbations are evaluated at different positions given by Eq. (47). In evaluating the branches of the tree, we encounter products of the form

$$\exp(-s_1 iKv\tau')\exp[-s_2 iKv(\tau'+\tau'')] \times \exp[-s_3 iKv(\tau'+\tau''+\tau''')], \tag{59}$$

where the signs s_n are associated with perturbations occuring at t', t'', and t''' for s_1, s_2, and s_3, respectively, and are given by

$$s_n = \pm \text{ for } d_\mu \text{ or } d_\sigma = \pm 1 \\ = \mp \text{ for } d_\rho = \pm 1 . \tag{60}$$

Table I summarizes the calculations for all 32 branches, where we have indicated only the sign on d_μ, d_ρ, and d_σ. Eight of these are set to zero since they lead to rapidly varying terms in space ($e^{\pm i3Ks}$). The tenth column indicates to which traveling wave the term contributes. The times of perturbation (and hence the place) associated with μ, ρ, and σ are given for each group of eight in column one. An expansion of the $t=1$ branch in terms of $V^r_{a'b'}$ and $V^l_{a'b'}$ is given in Appendix A. The T_{tw} column indicates which T integral is involved and will be discussed shortly.

TABLE I. Contributions of each branch of the perturbation tree of Fig. 3. The directions d_μ, d_ρ, and d_σ have been indicated by a plus (+ 1) and minus (− 1) sign for simplicity. The signs s_n are defined in Eqs. (59) and (60). The subscript w for the T_{tw} integrals may be calculated from the signs s_n as given in Eq. (69). Each row contributes to the complex polarization of the traveling wave with the perturbation as shown in the t column.

t	d_μ	d_ρ	d_σ	s_1	s_2	s_3	w	T_{tw}	d_n
	+	+	+	+	−	+	2	T_{12}	−
	+	+	−	+	−	−	3	T_{13}	+
1	+	−	+	+	+	+		0	
	+	−	−	+	+	−	1	T_{11}	−
$\mu \longleftrightarrow t'$	−	+	+	−	−	+	1	T_{11}	+
$\rho \longleftrightarrow t''$	−	+	−	−	−	−		0	
$\sigma \longleftrightarrow t'''$	−	−	+	−	+	+	3	T_{13}	−
	−	−	−	−	+	−	2	T_{12}	+
	+	+	+	+	+	−	1	T_{21}	−
	+	+	−	+	−	−	3	T_{23}	+
2	+	−	+	+	+	+		0	
	+	−	−	+	−	+	2	T_{22}	−
$\mu \longleftrightarrow t'$	−	+	+	−	+	−	2	T_{22}	+
$\rho \longleftrightarrow t'''$	−	+	−	−	−	−		0	
$\sigma \longleftrightarrow t''$	−	−	+	−	+	+	3	T_{23}	−
	−	−	−	−	−	+	1	T_{21}	+
	+	+	+	+	+	−	1	T_{31}	−
3	+	+	−	−	+	−	2	T_{32}	+
	+	−	+	+	+	+		0	
$\mu \longleftrightarrow t''$	+	−	−	−	+	+	3	T_{33}	−
$\rho \longleftrightarrow t'''$	−	+	+	+	−	−	3	T_{33}	+
$\sigma \longleftrightarrow t'$	−	+	−	−	−	−		0	
	−	−	+	+	−	+	2	T_{32}	−
	−	−	−	−	−	+	1	T_{31}	+
	+	+	+	+	−	+	2	T_{42}	−
4	+	+	−	−	−	+	1	T_{41}	+
	+	−	+	+	+	+		0	
$\mu \longleftrightarrow t'''$	+	−	−	−	+	+	3	T_{43}	−
$\rho \longleftrightarrow t''$	−	+	+	+	−	−	3	T_{43}	+
$\sigma \longleftrightarrow t'$	−	+	−	−	−	−		0	
	−	−	+	+	+	−	1	T_{41}	−
	−	−	−	−	+	−	2	T_{42}	+

Appropriately multiplying and summing the terms of the tree, as discussed in Sec. IV, to form $\rho^{(3)}_{a'b'}$ and substituting this result into Eq. (37), one has the third-order contribution to the complex polarization:

$$\mathcal{P}^{(3)}_n(t) = (\hbar^3 Ku)^{-1} \sum_\mu \sum_\rho \sum_\sigma E_\mu E_\rho E_\sigma e^{i\psi_{n\mu\rho\sigma}} \sum_{a'} \sum_{b'} \sum_{a''} \sum_{b''} \delta_{a',b'+p_n}\delta_{a'',b'+p_\mu}\delta_{b'',a''-p_\rho}\delta_{a',b''+p_\sigma} \times \mathcal{P}_{b'a'}\mathcal{P}_{a''b'}\mathcal{P}_{b''a''}\mathcal{P}_{a'b''} \sum_{t=1}^{4} \mathfrak{T}_{t,w(d_\mu,d_\rho,d_\sigma,t)} , \tag{61}$$

where the relative phase angle

$$\psi_{n\mu\rho\sigma} = (\nu_n - \nu_\mu + \nu_\rho - \nu_\sigma)t + \phi_n - \phi_\mu + \phi_\rho - \phi_\sigma . \tag{62}$$

The $\mathfrak{T}_{t,w(d_\mu,d_\rho,d_\sigma,t)}$ are integrals of the form

$$T_{tw} = iKN_{tw}\pi^{-1/2}\int_{-\infty}^{\infty} dv\, e^{-(v/u)^2} \times \int_0^\infty d\tau' \int_0^\infty d\tau'' \int_0^\infty d\tau''' \exp\{-[(v_{t1}+iC_{w1}Kv)\tau' + (v_{t2}+iC_{w2}Kv)\tau'' + (v_{t3}+iC_{w3}Kv)\tau''']\}, \tag{63}$$

where the N_{tw} are defined in Table II and the v_{tk} in Table III. The subscript w is dependent on the directions d_μ, d_ρ, and d_σ via signs s_n:

$$\begin{aligned} w &= 1 \text{ for } s_1 = s_2 = -s_3 \\ &= 2 \text{ for } s_1 = -s_2 = s_3 \\ &= 3 \text{ for } s_1 = -s_2 = -s_3 . \end{aligned} \tag{64}$$

The C_{tw} are defined by the matrix

$$C_{tw} = \begin{pmatrix} -1 & 0 & 1 \\ 1 & 0 & 1 \\ 1 & 2 & 1 \end{pmatrix} . \tag{65}$$

The T_{tw} integrals may be expressed in terms of the plasma dispersion function of Eq. (53), as discussed in Appendix B. For later convenience we define

$$v_1 \equiv v_{11} = v_{21} = v_{31} = v_{41}, \quad v_2 \equiv v_{12} = v_{22}, \quad v_3 \equiv v_{32} = v_{42}; \tag{66}$$

$$v_4 \equiv v_{13}, \quad v_5 = v_{23} = v_{33}, \quad v_6 \equiv v_{43} . \tag{67}$$

The $\sum_{t=1}^{4}\mathfrak{T}_{t,w,(d_\mu,d_\rho,d_\sigma,t)}$ may be expressed as three types of integrals. With the use of the definitions in Eq. (67), this is summarized in Table IV, along with "strong-Doppler-limit" [$Ku \gg \gamma$ (see Appendix B)] form of the integrals. With the help of Eq. (B10) of Appendix B, the sum of T_{tw}

TABLE II. Definition of the N_{tw} which appear in the third-order integrals of Eq. (63) in terms of the $N_{a',b'}$ which are defined in Eq. (54).

N_{tw}	$w=1$	2	3
$t=1$	$N_{a'b'',d_n(n-\mu-\rho+\sigma)}$	$N_{a'b'',d_n(n-\mu+\rho-\sigma)}$	$N_{a'b'',d_n(n+\mu-\rho-\sigma)}$
2	$N_{b''a'',d_n(n-\mu+\rho-\sigma)}$	$N_{b''a'',d_n(n-\mu-\rho+\sigma)}$	$N_{b''a'',d_n(n+\mu-\rho-\sigma)}$
3	$N_{b''a'',d_n(n-\mu+\rho-\sigma)}$	$N_{b''a'',d_n(n+\mu-\rho-\sigma)}$	$N_{b''a'',d_n(n-\mu-\rho+\sigma)}$
4	$N_{a''b',d_n(n+\mu-\rho-\sigma)}$	$N_{a''b',d_n(n-\mu+\rho-\sigma)}$	$N_{a''b',d_n(n-\mu-\rho+\sigma)}$

for a homogeneously broadened medium, where $u \to 0$, is given by

$$T_{1i} + T_{2j} + T_{3k} + T_{4l} = i\left(\frac{Ku}{v_1}\right) \times \left[\frac{1}{v_2}\left(\frac{N_{1i}}{v_4} + \frac{N_{2j}}{v_5}\right) + \frac{1}{v_3}\left(\frac{N_{3k}}{v_5} + \frac{N_{4l}}{v_6}\right)\right] . \tag{68}$$

VII. AMPLITUDE- AND FREQUENCY-DETERMINING EQUATIONS

Substituting the real and imaginary parts of Eq. (61) for the third-order complex polarization into the self-consistency equations (15) and adding this contribution to Eq. (56), one has the amplitude- and frequency-determining equations to third order:

$$\dot{E}_n = \mathrm{Im}\left(\sum_\mu \bar{\alpha}_{n\mu}E_\mu e^{i\psi_{n\mu}} - \sum_\mu\sum_\rho\sum_\sigma \bar{\theta}_{n\mu\rho\sigma}E_\mu E_\rho E_\sigma e^{i\psi_{n\mu\rho\sigma}}\right), \tag{69}$$

$$\nu_n + \dot{\phi}_n = \Omega_n + E_n^{-1}\,\mathrm{Re}\left(\sum_\mu \bar{\alpha}_{n\mu}E_\mu e^{i\psi_{n\mu}} - \sum_\mu\sum_\rho\sum_\sigma \bar{\theta}_{n\mu\rho\sigma}E_\mu E_\rho E_\sigma e^{i\psi_{n\mu\rho\sigma}}\right), \tag{70}$$

where the complex saturation coefficients are

TABLE III. Definition of the complex frequencies v_{tk} used in the third-order integrals of Eq. (63).

v_{tk}	$k=1$	2	3
$t=1$	$\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu+\nu_\rho-\nu_\sigma)$	$\gamma_{a'a''}+i(\omega_{a'a''}+\nu_\rho-\nu_\sigma)$	$\gamma_{a'b''}+i(\omega_{a'b''}-\nu_\sigma)$
2	$\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu+\nu_\rho-\nu_\sigma)$	$\gamma_{a'a''}+i(\omega_{a'a''}+\nu_\rho-\nu_\sigma)$	$\gamma_{b''a''}+i(\omega_{b''a''}+\nu_\mu)$
3	$\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu+\nu_\rho-\nu_\sigma)$	$\gamma_{b''b'}+i(\omega_{b''b'}+\nu_\rho-\nu_\mu)$	$\gamma_{b''a''}+i(\omega_{b''a''}+\nu_\mu)$
4	$\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu+\nu_\rho-\nu_\sigma)$	$\gamma_{b''b'}+i(\omega_{b''b'}+\nu_\rho-\nu_\mu)$	$\gamma_{a''b'}+i(\omega_{a''b'}-\nu_\mu)$

$$\vec{\theta}_{n\mu\rho\sigma}=\frac{1}{2}(\hbar^3 Ku\epsilon_0)^{-1}\sum_{a'}\sum_{b'}\sum_{a''}\sum_{b''}\delta_{a',b'+p_n}\delta_{a'',b'+p_\mu}\delta_{b'',a''-p_\rho}\delta_{a',b''+p_\sigma}\varphi_{b'a'}\varphi_{a''b'}\varphi_{b''a''}\varphi_{a'b''}\sum_{t=1}^{4}\mathfrak{T}_{t,w(d_\mu,d_\rho,d_\sigma,t)}. \tag{71}$$

The remaining symbols are defined by the equations indicated: $\bar{\alpha}_{n\mu}$ [(58)]; $\phi_{n\mu}$ [(52)]; $\psi_{n\mu\rho\sigma}$ [(64)]; p_n [(28)]; Ω_n [(14)]; $\varphi_{a'b'}$ [(26) and (27)]; $\mathfrak{T}_{t,w}$ [Tables II–IV and (65)].

General amplitude- and frequency-determining equations taking into account arbitrarily oriented magnetic field, isotopes, general basis vectors, and hyperfine structure are given in Appendix C in extension of the theory of Sargent *et al.*[1]

VIII. MULTIMODE UNIDIRECTIONAL THEORY

The reduction to a unidirectional ring laser is accomplished by requiring $d_n=d_\mu=d_\rho=+1$ for traveling waves in the $+s$ direction. Since n, μ, ρ, and σ no longer index traveling waves, there is no backscattering and the $\vec{G}$ matrix [Eq. (12)] reduces to an anisotropy matrix for different polarizations, as in Ref. 1, Sec. II. Specifically, the first-order $\mu\rightarrow n'=ni'$, where, as in Sec. II, i' indexes polarizations and now n indexes only Fox-Li quasimodes. In the third order, only terms with $d_\mu-d_\rho=d_\sigma$ in Table I survive in the unidirectional limit. This results in considerable simplification since the only remaining integrals are those of type I in Table IV.

The amplitude- and frequency-determining equations (69) and (70) become

$$\dot{E}_n=\mathrm{Im}\left(\sum_{i'}\bar{\alpha}_{nn'}E_n e^{i\psi_{nn'}}-\sum_{\mu}\sum_{\rho}\sum_{\sigma}\bar{\theta}_{n\mu\rho\sigma}E_\mu E_\rho E_\sigma e^{i\psi_{n\mu\rho\sigma}}\right), \tag{72}$$

$$\nu_n+\dot{\phi}_n=\Omega_n+E_n^{-1}\,\mathrm{Re}\left(\sum_{i'}\bar{\alpha}_{nn'}E_{n'}e^{i\psi_{nn'}}-\sum_{\mu}\sum_{\rho}\sum_{\sigma}\bar{\theta}_{n\mu\rho\sigma}E_\mu E_\rho E_\sigma e^{i\psi_{n\nu\rho\sigma}}\right), \tag{73}$$

where Ω_n is the resonant frequency of the rotating cavity, n, μ, ρ, and σ index only Fox-Li quasimodes and polarizations, $\bar{\alpha}_{nn'}$ is given by Eq. (58) without the d_n on the $N_{a'b'}$, and

$$\vec{\theta}_{n\mu\rho\sigma}=\frac{1}{2}\nu(\hbar^3 Ku\epsilon_0)^{-1}\sum_{a'}\sum_{b'}\sum_{a''}\sum_{b''}\delta_{a',b'+p_n}\delta_{a'',b'+p_\mu}\delta_{b',a'-p_\rho}\delta_{a',b''+p_\sigma}\times\varphi_{b'a'}\varphi_{a'b'}\varphi_{b''a''}\varphi_{a'b''}\sum_{t=1}^{4}T_{t,w(t)}, \tag{74}$$

where $\sum_{t=1}^{4}T_{t,w(t)}$ is a type-I integral defined in Table IV.

Equation (74) may be easily expressed in terms of the strong Doppler limit discussed in Appendix B. Using the form given in Table IV for a type-I integral in this limit and noting from Table II that $N_{21}=N_{31}$, one has

$$\vec{\theta}_{n\mu\rho\sigma}=i\frac{1}{2}\nu\pi^{1/2}(\hbar^3 Ku\epsilon_0)^{-1}\sum_{a'}\sum_{b'}\sum_{a''}\sum_{b''}\delta_{a',b'+p_n}\delta_{a'',b'+p_\mu}\delta_{b'',a''-p_\rho}\delta_{a',b''+p_\sigma}\times\varphi_{b'a'}\varphi_{a''b'}\varphi_{b''a''}\varphi_{a'b''}N_{b''a''}(t)\mathfrak{D}(\tfrac{1}{2}\omega_{a'b'}+\tfrac{1}{2}\omega_{b''a''}-\tfrac{1}{2}\nu_\mu+\nu_\rho-\tfrac{1}{2}\nu_\sigma)[\mathfrak{D}(\omega_{a'a''}+\nu_\rho-\nu_\sigma)+\mathfrak{D}(\omega_{b''b'}+\nu_\rho-\nu_\mu)], \tag{75}$$

TABLE IV. Three distinct types of the above integrals are given in this table as sums of the T_{tw} integrals. If $Ku\gg\gamma$, the "strong Doppler limit" form may be used (see Appendix B). The v_i are defined in Eq. (67).

	$d_\mu\ d_\rho\ d_\sigma$	$\sum_t \mathfrak{T}_{t,w(d_\mu,d_\rho,d_\sigma)}$	Strong-Doppler-Limit Form
Type I	$\pm\ \pm\ \pm$	$T_{12}+T_{21}+T_{31}+T_{42}$	$T_{21}+T_{31}=(i2\pi^{1/2})(N_{21}v_2^{-1}+N_{31}v_3^{-1})(v_1+v_5)^{-1}$
Type II	$\pm\ \pm\ \mp$	$T_{13}+T_{23}+T_{32}+T_{41}$	$T_{41}=(i2\pi^{1/2})N_{41}[v_3(v_1+v_6)]^{-1}$
Type III	$\pm\ \mp\ \mp$	$T_{11}+T_{22}+T_{33}+T_{43}$	$T_{11}=(i2\pi^{1/2})N_{11}[v_2(v_1+v_4)]^{-1}$

where, for example,

$$\mathfrak{D}(\omega_{a'a''}+\nu_\rho-\nu_\sigma)=[\gamma_{a'a''}+i(\omega_{a'a''}+\nu_\rho-\nu_\sigma)]^{-1} \tag{76}$$

and $N_{b''a''}(t)$ is N_{21} (or N_{31}) of Table II and is given by

$$N_{b''a''}(t)=L^{-1}\int_0^L ds N_{b''a''}(s,t) \times\exp[i(K_n-K_\mu+K_\rho-K_\sigma)s]. \tag{77}$$

Tables II and III may still be used to define the N_{tw} and v_{tk} involved in the T_{tw} integrals, provided one remembers that there is no summation over traveling waves. The correspondence is possible due to the generality of the perturbation tree of Fig. 3. The perturbation tree for the unidirectional ring laser has no subbranches similar to the previous case (see Appendix A), and is given directly by Fig. 3 with the appropriate substitutions for the perturbations $V_{a'b'}$.

IX. DISCUSSION

A portion of the work concerning the extension of the scalar theory to the ring-laser configuration has been done by Aronowitz[8] and Aronowitz and Collins.[10,11] More recent treatments have been given by Menegozzi and Lamb[3] and O'Bryan and Sargent.[12] The latter treatments have employed a consistent representation of the electric field and use the electromagnetic field equations for a frame in uniform rotation derived by Irvine.[4] Menegozzi and Lamb[3] included backscattering and localized loss effects by permitting "bumps," steep space variations in the linear electric susceptibility, and O'Bryan and Sargent[12] considered multimode operation.

Our treatment has included backscattering and anisotropies in a general anisotropy matrix. Further, we have allowed for multimode electric fields with arbitrary polarization and, specifically, two circular polarizations, whereas previous work has concentrated on linear polarization.

Extensive discussion of previous papers dealing with laser oscillators subject to dc magnetic fields is given by Sargent *et al.*[1] in Sec. II. The extension of the scalar theory to include a dc magnetic field, atoms with hyperfine structure and arbitrary angular momenta, and isotropic abundance given by Heer and Graft[13] is general enough to describe the ring-laser case given here. However, they did not evaluate any of the coefficients in their amplitude- and frequency-determining equations (or in any special cases). They also assumed cavity losses to be isotropic and neglected backscattering. Fradkin and Khayutin[14] outlined a calculation similar to the four-mode discussion in our forthcoming paper.[15] They obtained similar coupling coefficients; however, their expressions differ in that they are of order $(\gamma_{ab}/Ku)^2$. This results from a higher-order expansion of the plasma dispersion function[16] in the "Doppler limit." The coupling coefficients derived here may be calculated exactly using the exact expressions for the T_{tw} integrals given in Appendix A.

The present calculation gives the amplitude- and frequency-determining equations for the multimode operation of a ring laser subject to an axial dc magnetic field with possible anisotropic and backscattering losses, and resonance and an active medium consisting of atoms which may have arbitrary angular momenta. Similar equations which include hyperfine structure, arbitrarily oriented magnetic field, and isotropic abundance have been given in Appendix C. The special cases of bidirectional and unidirectional operation for a limited number of waves will be given in a second paper[15] with graphs of coupling parameters versus J value of the upper state and magnetic field strength.

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APPENDIX A: EXPANSION OF $t=1$ BRANCH OF PERTURBATION TREE

Expressing the perturbation $V_{a'b'}$ as the sum of a right and left traveling wave, we write

$$V_{a'b'}=V^r_{a'b'}+V^l_{a'b'}. \tag{A1}$$

With this form, each branch in Fig. 3 yields eight subbranches. Expanding the $t=1$ branch we obtain Fig. 4. Employing the subscript associations of Sec. VI, using Eq. (46) and definition (48), we find for the leftmost branch of Fig. 4

$$-\left(\frac{i}{\hbar^3}\right)W(v)\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\exp[-(\gamma_{a'b'}+i\omega_{a'b'})\tau']\sum_{a''}\sum_{b''}V^r_{a''b'}(s',t') \times\exp[-(\gamma_{a'a''}+i\omega_{a'a''})\tau'']V^r_{b''a''}(s'',t'')\exp[-(\gamma_{a'b''}+i\omega_{a'b''})\tau''']V^r_{a'b''}(s''',t''')N_{a'b''}(s,t). \tag{A2}$$

Noting that the explicit form for $V^r_{a'b'}$ contains e^{iKs}, using Eq. (47), and interchanging the order of summa-

tion and integration, one has

$$-i(2^{1/2}\hbar)^{-3}W(v)\sum_{\mu}\sum_{\rho}\sum_{\sigma}E_\mu E_\rho E_\sigma \exp\{-i[(\nu_\mu-\nu_\rho+\nu_\sigma)t+\phi_\nu-\phi_\rho-\phi_\sigma]\}$$

$$\times\sum_{a''}\sum_{b''}\delta_{a'',b'+p_\mu}\delta_{b'',a''-p_\rho}\delta_{a',b''+p_\sigma}\mathcal{P}_{a''b'}\mathcal{P}_{b''a''}\mathcal{P}_{a'b''}$$

$$\times\exp[i(K_\mu-K_\rho+K_\sigma)s]\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\exp[-iKv(\tau'+\tau''')]$$

$$\times\exp\{-[\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu+\nu_\rho-\nu_\sigma)]\tau'\}$$

$$\times\exp\{-[\gamma_{a'a''}+i(\omega_{a'a''}+\nu_\rho-\nu_\sigma)]\tau''\}\exp\{-[\gamma_{a'b''}+i(\omega_{a'b''}-\nu_\sigma)]\tau'''\}, \tag{A3}$$

where for this branch $d_\mu=d_\rho=d_\sigma=+1$ and $s_1=-s_2=s_3=+1$, in agreement with Table I. The remaining branches may be calculated in a similar manner.

APPENDIX B: INTEGRATION OF THIRD-ORDER INTEGRALS

The third-order integrals

$$T_{tw}=iK^{-1/2}N_{tw}\int_{-\infty}^{\infty}dv\exp[-(v/u)^2]\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\exp\{-[(v_1+iC_1Kv)\tau'+(v_2+iC_2Kv)\tau''+(v_3+iC_3Kv)\tau''']\} \tag{B1}$$

may be evaluated in terms of the plasma dispersion function of Eq. (53) with use of a simple partial-fraction expansion. Here, for typographical simplicity, we use v_k for v_{tk} and C_k for C_{wk}. The time integrations in Eq. (B1) may be performed immediately, yielding

$$T_{tw}=iK\pi^{-1/2}N_{tw}\int_{-\infty}^{\infty}dv\,e^{-(v/u)^2}[(v_1+iC_1Kv)\times(v_2+iC_2Kv)(v_3+iC_3Kv)]^{-1}. \tag{B2}$$

Expanding the term in square brackets by partial fractions, we find

$$[(v_1+iC_1Kv)(v_2+iC_2Kv)(v_3+iC_3Kv)]^{-1}=\left(\frac{1}{C_1v_2-C_2v_1}\right)\left[\left(\frac{C_1}{C_1v_3-C_3v_1}\right)\left(\frac{C_1}{v_1+iC_1Kv}-\frac{C_3}{v_3+iC_3Kv}\right)-\left(\frac{C_2}{C_2v_3-C_3v_2}\right)\left(\frac{C_2}{v_2+iC_2Kv}-\frac{C_3}{v_3+iC_3Kv}\right)\right]; \tag{B3}$$

with the definition (53) and Eq. (B3), T_{tw} becomes

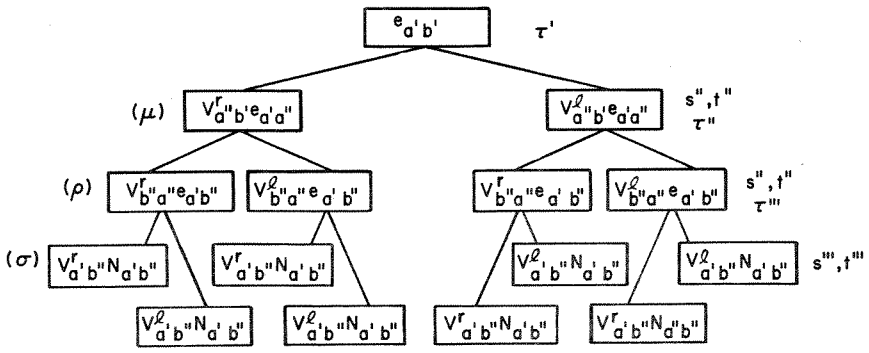

FIG. 4. $t=1$ branch of the perturbation tree in Fig. 3 is further expanded here.

$$T_{tw}=N_{tw}\left(\frac{1}{C_1v_2-C_2v_1}\right)\left[C_1\left(\frac{Z(v_1/C_1)-Z(v_3/C_3)}{C_1v_3-C_3v_1}\right)-C_2\left(\frac{Z(v_2/C_2)-Z(v_3/C_3)}{C_2v_3-C_3v_2}\right)\right]. \tag{B4}$$

We may now substitute the definitions of C_{wk} from Eq. (65) into Eq. (B4):

$$T_{t1}=\frac{(N_{t1}/v_2)[Z(v_1)+Z(v_3)]}{v_1+v_3}, \tag{B5}$$

$$T_2=\frac{(N_{t2}/v_3)[Z(v_1)-Z(v_3)]}{v_3-v_1}, \tag{B6}$$

and

$$T_{t3}=\frac{1}{2}\left(\frac{N_{t3}}{v_1-\frac{1}{2}v_2}\right)\left(\frac{Z(v_3)-Z(v_1)}{v_3-v_1}-\frac{Z(v_3)-Z(\frac{1}{2}v_2)}{v_3-\frac{1}{2}v_2}\right). \tag{B7}$$

If $v_1\to v_3$, both the numerators and denominators of the terms involving (v_1-v_3) in Eqs. (B6) and (B7) approach zero. In this limit, these terms become derivatives with respect to v_1, yielding

$$T_{t2}=\frac{(N_{t2}/v_3)\,dZ(v_1)}{dv_1} \tag{B8}$$

and

$$T_{t3}=N_{t3}(v_1-\tfrac{1}{2}v_2)^{-1}\left(\frac{dZ(v_1)}{dv_1}-\frac{Z(v_1)-Z(\frac{1}{2}v_2)}{v_3-\frac{1}{2}v_2}\right). \tag{B9}$$

As the atomic velocity $u\to 0$, $Z(v)\to i(Ku/v)$, and (B5)–(B7) reduce to

$$T_{tw}\to iKuN_{tw}/(v_1v_2v_3). \tag{B10}$$

In the case of large Doppler broadening where $Ku\gg$ decay rates and beat frequencies, $Z(v)\to i\pi^{1/2}$ and T_{tw} reduces to the "strong Doppler limit"

$$T_{t1}\to 2i\pi^{1/2}N_{t1}(v_1+v_3)^{-1},\quad T_{t2}\sim T_{t3}\sim 0. \tag{B11}$$

APPENDIX C: GENERAL AMPLITUDE AND FREQUENCY EQUATIONS

To consider general amplitude- and frequency-determining equations involving an arbitrarily oriented magnetic field, isotopes, general basis vectors, and hyperfine structure, we follow the treatment of Sargent *et al.*[1] As discussed in Sec. II of that reference, we assume that the jth isotope of the active medium has nuclear spin I_j, most probable $\hat{s}$ component of velocity u_j, and a fractional abundance a_j properly normalized, such that

$$\sum_j a_j=1, \tag{C1}$$

where j indexes the isotopes present. We also assume that the magnetic field does not break down the coupling between I_j and J (F is a good quantum number). In the derivation of the perturbation energy $V_{a'b'}$ we introduce direction cosines $f_q(p_\mu)$ given by

$$f_{\pm1}(p_\mu)=\hat{e}(p_\mu)\cdot(\hat{i}\mp i\hat{j}'),\quad f_0(p_\mu)=\hat{e}(p_\mu)\cdot\hat{k}', \tag{C2}$$

where, as in our treatment, p_μ indexes polarizations, $e(p_\mu)$ are the general basis, and $\hat{j}'$ and $\hat{k}'$ are unit vectors along the s and y directions of the magnetic field (see Fig. 1 of Ref. 1). The matrix elements are given by Eq. (27) with J_a and J_b replaced by F_a and F_b, respectively. The general amplitude- and frequency-determining equations for a Zeeman ring laser corresponding to those in Ref. 1 are our equations (72) and (73) with the general complex first-order coefficients

$$\bar{\bar{\alpha}}_{n\mu}=-\frac{1}{2}i\nu g_{n\mu}+\nu(\hbar K\epsilon_0)^{-1}\sum_j\left(\frac{a_j}{u_j}\right)\sum_{k=-1}^{1}f_k(p_n)^*f_k(p_\mu)\sum_{a'}\sum_{b'}\delta_{a',b'+k}|\wp_{a'b'}|^2$$
$$\times N_{a',b',d_n(n-\mu)}Z[\gamma_{a'b'}+i(\omega_{a'b'}-\nu_\mu)], \tag{C3}$$

and the complex saturation coefficients

$$\bar{\bar{\theta}}_{n\mu\rho\sigma}=\nu(\hbar^3K\epsilon_0)^{-1}\sum_j\left(\frac{a_j}{u_j}\right)\sum_{k=-1}^{1}f_k(p_n)^*\sum_{q=-1}^{1}f_q(p_\mu)\sum_{r=-1}^{1}f_r(p_\rho)^*\sum_{s=-1}^{1}f_s(p_\sigma)$$
$$\times\sum_{a'}\sum_{b'}\sum_{a''}\sum_{b''}\delta_{a',b'+k}\delta_{a'',b'+q}\delta_{b'',a''-r}\delta_{a',b''+s}$$
$$\times\wp_{b'a'}\wp_{a''b'}\wp_{b''a''}\wp_{a'b''}\sum_{t=1}^{4}\mathfrak{T}_{t,w(d_\mu,d_\rho,d_\sigma,t)}. \tag{C4}$$

Tables II–IV may still be used to determine the $\mathfrak{T}_{t,w}$ provided one now subscripts the resonant frequencies $\omega_{a'b'}$, K_n, and matrix elements $\mathcal{P}_{a'b'}$ with the isotope index j, which we have refrained from including for typographical simplicity. The procedure for expressing the coefficients in terms of the relative excitation may be found in Sec. V of Ref. 1.

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