

Semiclassical theory of multimode operation in lasers: The high-intensity field solution

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The semiclassical interaction of a two-level system with a multimode electromagnetic field is discussed in detail. The equations of motion for the macroscopic density matrix are transformed into a single integral equation for the population density, with a complex kernel which can take into account both running- and standing-wave configurations. The iterative solution to this integral equation, which corresponds to an expansion in terms of powers of the electromagnetic field amplitudes, turns out to be divergent when the field amplitudes are greater than some critical value. In the case in which the kernel is a periodic or quasiperiodic function of time, a general solution to the integral equation is found, and this solution turns out to be unique and convergent for every value of the electromagnetic (e.m.) field amplitude. This solution is expressed through a continued-fraction expansion, whose terms are matrices, which can be readily obtained from the kernel. This solution is a generalization of the continued-fraction expansion usually met in the strong-signal semiclassical theory of lasers. Some particular cases are then treated in detail; among them, the solution to the multimode operation in laser devices is given in which the excited level lifetime is long compared with the period of the beating term between two adjacent modes. Although the theory presented is directed towards the steady-state regime in multimode lasers, it may also treat the transient regime of laser operations, or it may be extended to include phenomena where a strong e.m. field, made up of several modes equally spaced in frequency, interacts with a two-level system.

I. INTRODUCTION

In 1964 a fundamental paper¹ by Lamb initiated a series of works on the semiclassical theory of gas lasers. This theory proved very fruitful in describing a number of features of gas lasers, such as mode competition, the "pushing" and "pulling" of frequencies of the modes, mode locking, and detuning dip. It then was generalized by several authors² in order to include degeneracy of laser levels, vectorial treatment of the electromagnetic field in the cavity, laser operation in a static magnetic field (Zeeman laser), and unidirectional and bidirectional ring lasers. The semiclassical theory was therefore a powerful tool for investigating both single-mode and multimode operation, even if in the multimode case the calculations were very cumbersome.

All of the theory is based upon a perturbation expansion in a power series of the electromagnetic field, and in Lamb's paper the expansion was carried out to third order. Later, a number of authors³ made calculations of successive orders of the power series. Recently, a perturbation tree was developed by O'Bryan and Sargent,⁴ who described a suitable way of providing all the terms in the expansion by means of a graphical technique which enables one to make computer calculations of the series expansion easily.

However, because of its inherent characteristic of perturbative treatment, this formulation fails in predicting the behavior of the laser operation at a high level of excitation of the active medium.

This was first pointed out in a paper by Stenholm and Lamb,⁵ who developed a semiclassical theory suitable for treating high-intensity gas lasers. The divergence between the two treatments becomes already apparent at an excitation level \mathcal{X} as low as 1.1.⁵

The strong-signal theory was also able to explain some features in the experimental results of Bolwijn,⁶ for which the previous perturbative treatment was inadequate. The treatment of Stenholm and Lamb makes it possible to evaluate at all orders in the interaction energy the polarization and the inversion population density of the medium, when a single standing mode is operating in the laser cavity. When one allows for atomic velocity (Doppler broadening), the solution for both these quantities is given in terms of a continued fraction. In turn, this continued-fraction expansion proved quite a powerful tool in treating a large number of phenomena where the atomic-medium-electromagnetic-field interaction must be taken into consideration at all orders.⁷ The convergence properties of the continued-fraction expansion were discussed by Feldman and Feld.⁸ Since then, several authors⁹ have extended the strong signal theory in order to treat multimode phenomena in lasers.

The purpose of this paper is to discuss in detail a third approach to the interaction between a strong electromagnetic (multimode) field and an atomic two-level system. This method starts with an integral equation for the population inversion density; in the collisionless case, or in the

case where collisions do not affect the atomic motion, the integral equation may be put in a form known as Volterra's second-kind integral equation. This approach seems to have some advantages: It unifies the two theories discussed above, because one can obtain from it the expansion both in a power series of the electromagnetic field and in the continued fraction; also it allows one to estimate the radius of convergence of the series expansion, in some particular cases. Furthermore, its solution in terms of the continuant theory, in the single-mode case, suggests a generalization to multimode operation.

In two limiting cases, i.e., the close coupling of the modes and in the large-separation case, the integral equation may be solved, giving a good starting point for multimode analysis. In Sec. II the basic theory of the response of an atomic (two-level) system to a multimode electromagnetic field is given. In that section the integral equation is derived. In Sec. III the convergence of the series expansion which can be built from it is discussed, while Sec. IV is devoted to the solution of the equation with an infinite set of equations in infinite unknowns. It will be shown in the same section that the continued-fraction expansion, which is convergent, belongs to these solutions. In Sec. V the solution to the multimode case is presented; here it is shown that in most cases of physical interest the kernel of the integral equation is a periodic function of time, and therefore the inversion population density may be expanded in a Fourier series whose coefficients are determined by means of a matrix continued-fraction expansion. This expansion turns out to be convergent for any value of the electromagnetic field amplitude. Finally, an application of the theory to the stationary atoms case is presented in Sec. VI.

Our formalism can be applied to a number of problems where the interaction of a two-level system with a multimode electromagnetic (e.m.) field must be considered at all perturbative orders. For example, we can mention saturation spectroscopy, where the saturating beam and the probing beam are provided by lasers tuned at different frequencies.¹⁰

II. BASIC FORMULATION

The semiclassical theory of lasers is based upon a three-step procedure. First, one solves the Maxwell equations for the electromagnetic field in the cavity, with a source term given by the macroscopic polarization of the medium. Next, one evaluates the polarization of the medium induced by that field. Third, a self-consistency requirement is made: the field generated by the polarization must be the same field which induces

the polarization. However, in treating the coupled Maxwell-Bloch equations of motion, a simplifying assumption can be made. In most laser cavities, the modes of the electromagnetic field have sharp resonances with high values of the quality factor Q . On the other hand, the atomic lifetimes, i.e., the decay times of the inversion population density and of the polarization, are typically several orders of magnitude shorter than the lifetime of the electromagnetic field in the cavity. Thus the atomic variables are assumed to follow adiabatically the field variables in the equations describing the interaction between an e.m. field and matter. The field amplitude is assumed to be constant during the time in which the atomic variables change appreciably.

Because of this discrimination, the Maxwell equations for the field amplitudes and phases are readily written¹¹: the field variables of the n th mode depend only on that component of the polarization which is the projection of the macroscopic polarization on the n th mode.

Some problems arise when we consider the Bloch equations for matter. Here the broad line of gain of each atom allows several modes of the e.m. field to induce the component of the polarization in the n th mode. As a result of the high field intensities, nonlinear beatings of the modes are of importance, and an adequate treatment must consider them at all orders.

In what follows, we shall consider the Bloch equations of motion for matter, and we shall derive an integral equation for the inversion population density n , where the beating terms of the e.m. field couple the Fourier components of n .

We are primarily interested in the macroscopic polarization induced in an atomic medium (a gas with moving atoms) by a classical electromagnetic field $\vec{E}(\vec{r}, t)$ which is made up of a number of quasis-monochromatic components,

$$\vec{E}(\vec{r}, t) = \sum_n A_n(t) \vec{U}_n(\vec{r}). \quad (1)$$

Here, $A_n(t)$ has a rapid oscillation at frequency ν_n , and it may be modulated in time with characteristic frequencies much smaller than ν_n . $\vec{U}_n(\vec{r})$ represents the spatial variation of the field. The form (1) can represent both a standing-wave and a running-wave configuration. We shall distinguish between them at a later time.

Let us assume that all frequencies of the e.m. field are nearly resonant with a two-level (non-degenerate) transition of the atomic medium, so that only these two levels are involved in the interaction, while the other levels form a nonabsorptive background which may be taken into account by redefining the index of refraction of the medium.

Let $|a\rangle$ and $|b\rangle$, respectively, be the upper and the lower states of any atom of the medium, with energies E_a and E_b . The transition frequency ω is then given by

$$E_a - E_b = \hbar\omega. \quad (2)$$

We assume that the electric field in (1) propagates along one direction (say the z direction, which in laser theories is assumed to be the axis of the optical cavity), while its amplitude is constant in a plane perpendicular to that direction. With such assumptions, $\vec{U}_n(\vec{r}) = U_n(z)\hat{e}_x$, where \hat{e}_x is a unit vector perpendicular to the z axis.

A typical atom with an axial velocity v is excited by an external pumping mechanism to an incoherent superposition of levels a and b . The excitation takes place at a certain coordinate z_0 and at a time t_0 . If the atom does not suffer collisions which can change its velocity, at a later time t it will be in the position

$$z = z_0 + v(t - t_0). \quad (3)$$

Starting from t_0 , the atom interacts coherently with the e.m. field; in a reference frame which moves according to (3) the atom is at rest, and its density matrix obeys the equation of motion

$$i\hbar\dot{\rho} = [H, \rho] \quad (4)$$

starting from an initial value, at $t=t_0$, which, for the particular pumping mechanism, takes the form

$$\rho(t_0, t_0) = \begin{pmatrix} \lambda_a & 0 \\ 0 & \lambda_b \end{pmatrix}. \quad (5)$$

Equation (5) represents a single-atom initial condition. However, all atoms created at (z_0, t_0) with the same axial velocity v will have the same interaction with the electromagnetic field, or, in other words, will be represented by the same density matrix ρ whose time evolution is given by (4) and (5). We can therefore assume that all these atoms are described by (4) and (5). Then the initial condition (5) represents the density of atoms created at (z_0, t_0) in the upper (λ_a) or lower (λ_b) state, with an initial velocity v .

We assume that the pumping mechanism is homogeneous and constant in time, so that λ_a and λ_b will depend only on v . Furthermore, the v dependence is assumed to be the same for λ_a and λ_b , so that we can factorize the initial condition as

$$\rho(t_0, t_0) = W(v) \begin{pmatrix} \Lambda_a & 0 \\ 0 & \Lambda_b \end{pmatrix}.$$

We do not need to specialize $W(v)$ as a Maxwell

distribution of velocities for atoms, the treatment being quite general. We note that $W(v)$ may also include other mechanisms which cause nonhomogeneous broadening. In the stationary-atoms case, for example, nonhomogeneity may be induced by displacement of the transition frequency ω due to local strains in the crystal lattice in which the active atoms are embedded. In this case, $v=0$, and $W(v)$ must be replaced by the distribution of frequencies $W(\omega)$.

The Hamiltonian which appears in (4) is made up of two parts: the first one is the unperturbed Hamiltonian H_0 of the two-level atom,

$$H_0 = E_a |a\rangle\langle a| + E_b |b\rangle\langle b|, \quad (6)$$

while the second one is the interaction Hamiltonian H_1 which couples the atom to the field. We assume that the interaction is an electric dipole interaction. Furthermore, we assume that the dipole moment p_x of the atom has real matrix elements

$$\mathcal{P} = \langle a | p_x | b \rangle = \langle b | p_x | a \rangle, \quad (7)$$

$$\langle a | p_x | a \rangle = \langle b | p_x | b \rangle = 0. \quad (8)$$

With such assumptions, H_1 has the form

$$H_1 = -\mathcal{P}(|a\rangle\langle b| + |b\rangle\langle a|)E(z, t). \quad (9)$$

Equation (4) is then written for the four components of the density matrix:

$$\dot{\rho}_{aa} = -(i/\hbar)\mathcal{P}E(z, t)(\rho_{ab} - \rho_{ba}), \quad (10)$$

$$\dot{\rho}_{bb} = (i/\hbar)\mathcal{P}E(z, t)(\rho_{ab} - \rho_{ba}), \quad (11)$$

$$\dot{\rho}_{ab} = -i\omega\rho_{ab} - (i/\hbar)\mathcal{P}E(z, t)(\rho_{aa} - \rho_{bb}), \quad (12)$$

$$\rho_{ba} = \rho_{ab}^*. \quad (13)$$

The electric field as seen by the atom in the reference frame where it is at rest differs from the electric field as seen in the laboratory frame, and a proper Lorentz transformation should be performed in order to account for this change. However, the ratio of the atomic velocity v to the velocity of light c under usual experimental conditions is of the order of 10^{-6} , so that we shall confine ourselves to a nonrelativistic treatment. Therefore the electric field $E(z, t)$ is given, in the reference frame where the atom is at rest, by the relation

$$E(z, t) = E^{(+)}(z_0 + v(t - t_0), t) + E^{(-)}(z_0 + v(t - t_0), t), \quad (14)$$

where

$$E^{(+)}(z, t) = \frac{1}{2} \sum_n E_n(t) e^{-i(\nu_n t + \phi_n)} U_n(z), \quad (15)$$

$$E^{(-)}(z, t) = [E^{(+)}(z, t)]^*. \quad (16)$$

In (15), the summation runs over all modes into which the electric field has been decomposed. A

slow amplitude modulation can be allowed in $E_n(t)$, but in what follows we shall restrict ourselves to a steady-state field, so that the only time dependence of E comes from the optical oscillations, and from the variation of the z coordinate according to Eq. (3).

The spatial mode function $U_n(z)$ has different analytical expressions in a running-mode configuration or in a standing-mode configuration. In the former case, we assume it to vary according to

$$U_n(z) = e^{ik_n z}, \quad (17)$$

while in the latter case, $U_n(z)$ is a real function of z which we assume to be

$$U_n(z) = \sin k_n z. \quad (18)$$

A damping term is then introduced into Eqs. (10)–(12), in order to account for losses due to radiation damping or to some other relaxation mechanism. Equations (10)–(12) now read

$$\dot{\rho}_{aa} = -\gamma_a \rho_{aa} - (i\mathcal{P}/\hbar)E(z, t)(\rho_{ab} - \rho_{ba}), \quad (19)$$

$$\dot{\rho}_{bb} = -\gamma_b \rho_{bb} + (i\mathcal{P}/\hbar)E(z, t)(\rho_{ab} - \rho_{ba}), \quad (20)$$

$$\dot{\rho}_{ab} = -\gamma_{ab} \rho_{ab} - i\omega \rho_{ab} - (i\mathcal{P}/\hbar)E(z, t)(\rho_{aa} - \rho_{bb}). \quad (21)$$

This set of equations describes the time evolution of those atoms which an external pumping mechanism has created at time t_0 and at position z_0 , in an incoherent superposition of states $|a\rangle$ and $|b\rangle$. We can perform a formal integration of (19)–(21), obtaining the following set of integral equations:

$$\rho_{aa}(t, t_0) = -\frac{i\mathcal{P}}{\hbar} \int_{t_0}^t e^{-\gamma_a(t-t')} E(z_0 + v(t' - t_0), t') [\rho_{ab}(t', t_0) - \rho_{ba}(t', t_0)] dt' + \lambda_a e^{-\gamma_a(t-t_0)}, \quad (22)$$

$$\rho_{bb}(t, t_0) = \frac{i\mathcal{P}}{\hbar} \int_{t_0}^t e^{-\gamma_b(t-t')} E(z_0 + v(t' - t_0), t') [\rho_{ab}(t', t_0) - \rho_{ba}(t', t_0)] dt' + \lambda_b e^{-\gamma_b(t-t_0)}, \quad (23)$$

$$\rho_{ab}(t, t_0) = -\frac{i\mathcal{P}}{\hbar} \int_{t_0}^t e^{-(\gamma_{ab} + i\omega)(t-t')} E(z_0 + v(t' - t_0), t') [\rho_{aa}(t', t_0) - \rho_{bb}(t', t_0)] dt'. \quad (24)$$

In Eqs. (22)–(24), z_0 is a parameter. If we change it without changing t_0 or v , we choose another set of atoms which do not spatially overlap with the others, so that they contribute to the polarization at a different axial coordinate. Now we need to evaluate the polarization at the same time and at the same coordinate z , so that if we change z_0 , we must allow t_0 to change in order to allow all the atoms to be at the same position z at time t . This can be done by requiring that

$$z_0 + v(t' - t_0) = z - v(t - t'). \quad (25)$$

If we substitute Eq. (25) into (22)–(24), we obtain the integral equation for the density matrix ρ of those atoms which will be at the same position z at time t .

We can then add up contributions to the density matrix from all atoms, “selected” by the requirement (25), but created at any time t_0 prior to t . Thus we define a macroscopic density matrix

$$\rho^{(M)}(t, z, v) = \int_{-\infty}^t dt_0 \rho(t, t_0) \quad (26)$$

whose elements are given by

$$\rho_{aa}^{(M)}(t, z, v) = -\frac{i\mathcal{P}}{\hbar} \int_{-\infty}^t dt_0 \int_{t_0}^t dt' e^{-\gamma_a(t-t')} E(z - v(t - t'), t') [\rho_{ab}(t', t_0) - \rho_{ba}(t', t_0)] + \frac{\lambda_a}{\gamma_a}, \quad (27)$$

$$\rho_{bb}^{(M)}(t, z, v) = \frac{i\mathcal{P}}{\hbar} \int_{-\infty}^t dt_0 \int_{t_0}^t dt' e^{-\gamma_b(t-t')} E(z - v(t - t'), t') [\rho_{ab}(t', t_0) - \rho_{ba}(t', t_0)] + \frac{\lambda_b}{\gamma_b}, \quad (28)$$

$$\rho_{ab}^{(M)}(t, z, v) = -\frac{i\mathcal{P}}{\hbar} \int_{-\infty}^t dt_0 \int_{t_0}^t dt' e^{-(\gamma_{ab} + i\omega)(t-t')} E(z - v(t - t'), t') [\rho_{aa}(t', t_0) - \rho_{bb}(t', t_0)]. \quad (29)$$

The order of integration over t' and t_0 can be changed, and, using the fact that the electric field does not depend on t_0 , we get the following integral equations, in which there appear only elements of the macroscopic density matrix:

$$\begin{aligned} \rho_{aa}^{(M)}(t, z, v) - \rho_{bb}^{(M)}(t, z, v) = & -\frac{i\mathcal{P}}{\hbar} \int_{-\infty}^t dt' \{ (e^{-\gamma_a(t-t')} + e^{-\gamma_b(t-t')}) \dot{E}(z - v(t - t'), t') \\ & \times [\rho_{ab}^{(M)}(t', z', v) - \rho_{ba}^{(M)}(t', z', v)] \} + \frac{\lambda_a}{\gamma_a} - \frac{\lambda_b}{\gamma_b}, \end{aligned} \quad (30)$$

$$\rho_{ab}^{(M)}(t, z, v) = -\frac{i\mathcal{P}}{\hbar} \int_{-\infty}^t dt' e^{-(\gamma_{ab} + i\omega)(t-t')} E(z - v(t-t'), t') [\rho_{aa}^{(M)}(t', z', v) - \rho_{bb}^{(M)}(t', z', v)], \quad (31)$$

where z' is the coordinate of the atoms at the time t' , i.e.,

$$z' = z - v(t - t'). \quad (32)$$

In the following, we shall ignore the index (M) which characterizes the macroscopic density matrix, which will simply be indicated by $\rho(t, z, v)$.

We now substitute $\rho_{ab}(t, z, v)$, as given by (31), and its complex conjugate into (30), and find a single integral equation for the inversion population density,

$$n(t, z, v) = \rho_{aa}(t, z, v) - \rho_{bb}(t, z, v). \quad (33)$$

We get

$$n(t, z, v) = \frac{\lambda_a}{\gamma_a} - \frac{\lambda_b}{\gamma_b} - \frac{i\mathcal{P}}{\hbar} \int_{-\infty}^t dt' \int_{-\infty}^{t'} dt'' \left[(e^{-\gamma_a(t-t')} + e^{-\gamma_b(t-t')}) E(z - v(t-t'), t') E(z' - v(t' - t''), t'') \right. \\ \left. \times \left(-\frac{i\mathcal{P}}{\hbar} (e^{-(\gamma_{ab} + i\omega)(t'-t'')} + \text{c.c.}) [\rho_{aa}(t'', z'', v) - \rho_{bb}(t'', z'', v)] \right) \right], \quad (34)$$

where $z'' = z' - v(t' - t'') = z - v(t - t'')$.

Finally, we change the order of integration over t' and t'' in (34), obtaining the following integral equation for the inversion population density:

$$n(t, z, v) = N_0 - \frac{\mathcal{P}^2}{\hbar^2} \int_{-\infty}^t dt'' K(t, t'') \\ \times n(t'', z - v(t - t''), v). \quad (35)$$

The function $n(t'', z'', v)$ and the kernel $K(t, t'')$

$$K(t, t'') = E(z - v(t - t''), t'') \int_{t''}^t dt' E(z - v(t - t'), t') (e^{-\gamma_a(t-t')} + e^{-\gamma_b(t-t')}) (e^{-(\gamma_{ab} + i\omega)(t'-t'')} + \text{c.c.}). \quad (36)$$

In Appendix A the kernel (36) is evaluated for both running- and standing-wave configurations. Equation (35) is the integral equation for the inversion population density, and its derivation does not need any substantial approximation, because we did not use until now the fact that the e.m. field amplitude varies slowly in a time in which the atomic variables change appreciably. Thus it is applicable to a large class of problems in which one essentially deals with a two-level system (a spin- $\frac{1}{2}$ system) interacting with a classical multimode field, through an off-diagonal interaction Hamiltonian of the form (9). In what follows, we shall deal with a stationary regime for the electromagnetic field, and we shall search for stationary solutions of $n(t, z, v)$. But in view of the discussion

in the integrand of (35) depend on z , the former only through z'' and the latter also explicitly. Owing to the fact that the atoms do not change their velocity in the interaction time, it is possible to obtain an integral equation in which there is a functional dependence only on t . This will be discussed in Appendix A. In (35) $N_0 = \lambda_a/\gamma_a - \lambda_b/\gamma_b$ is the nonhomogeneous (source) term, i.e., the inversion population density in the absence of the e.m. field, and the kernel $K(t, t'')$ is given by

at the beginning of this section, these stationary solutions are capable of describing also the dynamics of the interaction process in a laser device. In fact, one immediately sees from (36) that the kernel $K(t, t'')$ depends on the values assumed by E in a time interval before t of the order of $1/\gamma_a$ ($\alpha = a, b$) or $1/\gamma_{ab}$. During this interval, the field amplitudes are assumed to be constant.

We note that knowledge of $n(t, z, v)$ is sufficient to determine, by integration of Eq. (31), the off-diagonal element of the density matrix ρ_{ab} . Then the macroscopic polarization of the atoms with velocity v along the z axis of the cavity is given by

$$P(z, v, t) = \mathcal{P} [\rho_{ab}(t, z, v) + \rho_{ba}(t, z, v)], \quad (37)$$

and an integration over the velocity distribution of atoms gives the polarization as a function of z and t :

$$P(z, t) = \int dv P(z, t, v). \quad (38)$$

In what follows, we shall focus our attention on the integral equation (35), which is the crucial point of the semiclassical theory of lasers.

III. CONVERGENCE OF THE ITERATIVE SOLUTION OF THE INTEGRAL EQUATION

In this section we shall briefly discuss the iterative solution to the integral equation (35). In order to simplify our discussion, we shall restrict ourselves to the simplest case, i.e., the situation where a single running wave is present. It is known that the steady-state solution of this case exists, and in the rotating-wave approximation it has a simple analytical form,¹² which we shall derive from the integral equation.

The kernel for this case is derived from (A5),

$$K(t, t'') = \sum_{\alpha=a,b} \frac{\frac{1}{4}E^2}{i(\nu - kv - \omega) + \gamma_\alpha - \gamma_{ab}} \times (e^{i(\nu - kv - \omega)(t' - t'')} e^{-\gamma_{ab}(t - t'')} - e^{-\gamma_\alpha(t - t'')}), \quad (39)$$

where we have dropped the index $n=1$. We note that K is independent of z , so that the solution to (35) will also be z independent. If we use the integral equation in the form (A9), we have

$$n(t, v) = N_0 - \frac{\mathcal{P}^2}{\hbar^2} \left(\sum_{\alpha=a,b} \frac{E^2}{4(\gamma_\alpha - \gamma_{ab} + i\Omega)} \times \int_0^\infty d\theta (e^{(i\Omega - \gamma_{ab})\theta} - e^{-\gamma_\alpha\theta}) \times n(t - \theta, v) + c.c. \right), \quad (40)$$

where $\Omega = \nu - kv - \omega$.

The fact that the kernel has no driving term dependent on t allows us to assume that a steady-state solution, independent of t , exists:

$$n(t, v) = n(v). \quad (41)$$

If we substitute (41) into (40), and perform the integration, we get

$$n(v) = \frac{N_0}{1 + (\mathcal{P}^2/\hbar^2)^{\frac{1}{2}} E^2 \gamma_{ab} (\gamma_a + \gamma_b) [\gamma_a \gamma_b (\gamma_{ab}^2 + \Omega^2)]^{-1}}, \quad (42)$$

which is the correct steady-state solution for one running-wave configuration. The population inversion density does not depend on z and t , but depends only on v , through Ω .

If we try to obtain the solution in an iterative

way, we find that when the saturation parameter

$$\sigma = \frac{\mathcal{P}^2}{\hbar^2} \frac{E^2}{2} \frac{\gamma_{ab}(\gamma_a + \gamma_b)}{\gamma_a \gamma_b} \frac{1}{\gamma_{ab}^2 + \Omega^2} \quad (43)$$

becomes greater than 1, the expansion diverges. This can be easily seen. Equation (40) is rewritten

$$n(t, v) = N_0 - \int_0^\infty d\theta f(\theta) n(t - \theta, v). \quad (44)$$

Then an iterative solution to (44) is obtained by inserting N_0 in place of $n(t - \theta, v)$ in the integrand of (44), obtaining just $n^{(1)}(t, v)$,

$$n^{(1)}(t, v) = N_0 - \int_0^\infty d\theta f(\theta) N_0 = N_0(1 - \sigma) \quad (45)$$

with

$$\sigma = \int_0^\infty d\theta f(\theta), \quad (46)$$

and then iterating this procedure. We get

$$n(t, v) = N_0(1 - \sigma + \sigma^2 - \sigma^3 + \dots), \quad (47)$$

which is convergent to the correct value (42) only if $\sigma < 1$, and otherwise is divergent.

We can ask why the solution diverges for σ values greater than 1. The integral equation (35) is a Volterra equation of the second kind, and the iterative procedure always gives the correct (convergent) solution for this kind of equation. But Eq. (35) is singular, because the lower limit of integration is $-\infty$. This singularity arises when we assume that the field amplitude has its stationary value at $t = -\infty$; when such a singularity is introduced in the integral equation, the convergence of the iterative solution is lost, at least for field intensities exceeding some critical values. In single-mode operation, such a critical value is reached when σ , as defined by (43), equals 1. For higher values of σ , one cannot obtain the steady-state value of the inversion population density $n(v)$ starting with an iterative procedure from the unperturbed value N_0 . The divergence of the expansion has already been discussed in the rate-equation formulation of gas laser theory.¹³ For multimode operation, a similar expansion is usually made in powers of the electric field intensities. This expansion can be performed directly from (35), substituting the function $n(t'', z'', v)$ in the integrand with N_0 ; then an integration yields $n^{(1)}(t, z, v)$, which in turn is put in the integrand in order to get $n^{(2)}(t, z, v)$, and so on. The so-called cubic approximation is equivalent to truncating this procedure after the first step, retaining just the E^2 dependence of

n and the E^3 dependence of the polarization. But also for the multimode case one can expect divergences. Thus the solution to Eq. (35) must be found in some other way, namely by assuming some steady-state analytical form for $n(t, z, v)$, just as was done above for the simplest case of single running-wave operation. To this kind of solution the following sections are devoted.

IV. INTEGRAL EQUATION FOR $n(t, z, v)$

As noted in Sec. III, the solution to the integral equation for $n(t, z, v)$ cannot be found, in the high-intensity field case, by means of an iterative procedure. We shall therefore describe a method, which proves to be convergent, for solving the integral equation, and present a generalization of the expansion in a continued fraction first used by Stenholm and Lamb⁵ in treating high-intensity laser theory.

The integral equation for $n(t, z, v)$ takes the form (A9)

$$n(t, z, v) = N_0 - \frac{\mathcal{P}^2}{\hbar^2} \sum_{p=1}^M A_p e^{i\Omega_p t} \times \int_0^\infty d\theta e^{\Gamma_p \theta} n(t - \theta, z, v). \quad (48)$$

From (48) we note that the driving terms in the kernel are oscillating at frequencies Ω_p which are the differences of any pair of optical frequencies of the modes shifted by the Doppler effect in a gas laser system. Thus we deduce that the inversion population density will oscillate with the same frequencies, and with their combinations. Owing to the form of (48), any oscillation in $n(t)$ at a certain frequency, say, ν , will influence directly the terms oscillating at frequencies $\nu + \Omega_p$; these in turn will influence the terms oscillating at $\nu + \Omega_p + \Omega_{p'}$, and so on. The source term in this chain is the average value of $n(t)$, which is a constant ($\nu = 0$). This suggests starting the chain with $\nu = 0$. A formal solution to Eq. (48) is then given by

$$n(t, z, v) = \sum_{\{k\}} B_{\{k\}}(z, v) \exp\left(\sum_{p=1}^M k_p \Omega_p t\right), \quad (49)$$

where the sum over $\{k\}$ means that we take any set of integer numbers $k_1, \dots, k_p, \dots, k_M$ ranging from 0 to ∞ . Since for any given value of Ω_p there is another p , say, \bar{p} , for which $\Omega_{\bar{p}} = -\Omega_p$, those sets $\{k\}$ in which $k_p = k_{\bar{p}}$ for any p will contribute to the average value of $n(t)$. At each step of the chain, there appear terms which contribute to the average value of $n(t)$. Thus in this form the problem seems to be rather intractable. However, there is a case in which the problem at

hand can be solved, and the solution is convergent. This special case is met when all the frequencies Ω_p are multiples of a fundamental frequency, say δ :

$$\Omega_p = m_p \delta, \quad (50)$$

where m_p is some integer, positive, negative, or zero. Equation (50) is not an unusual condition for the frequencies of oscillation of the inversion population density. For example, in a laser device in which the active atoms are fixed in some lattice sites, there is no Doppler shift, and condition (50) holds when each pair of adjacent modes are separated by a constant quantity

$$\delta = \pi c/L, \quad (51)$$

where L is the cavity length. In a (running-wave) unidirectional ring laser, the Doppler effect changes the frequencies by a factor which is identical for all the modes, so that, if the modes are separated by δ_0 ,

$$\delta = \delta_0(1 + v/c). \quad (52)$$

Furthermore, in the general case one could always choose a fundamental frequency δ such that all the frequencies Ω_p are multiples of δ , at least approximately.

Let us now turn to condition (50). If that is the case, we collect in the kernel of (48) all terms which have the same frequency Ω_p , and write the integral equation in the form

$$n(t, z, v) = N_0 - \frac{\mathcal{P}^2}{\hbar^2} \sum_{m=-N}^N e^{i m \delta t} \times \int_0^\infty d\theta f_m(\theta) n(t - \theta, z, v), \quad (53)$$

where we have used the fact that for each Ω_p there must exist a frequency $-\Omega_p$ in the kernel. In (53) $f_m(\theta)$ is made up of a sum of exponentials $e^{\Gamma_p \theta}$ (which correspond to the same frequency Ω_p) multiplied by the constant coefficients A_p . The sum runs from $-N$ to $+N$, and the condition that $n(t, z, v)$ is real assures us that

$$f_{-m}(\theta) = f_m^*(\theta). \quad (54)$$

The steady-state solution to (53) is simply found from considerations similar to those which lead to formula (49). Since there is a fundamental frequency δ such that each Ω_p is a multiple of that one, the solution to (53) is given by

$$n(t, z, v) = \sum_k n_k(z, v) e^{i k \delta t}. \quad (55)$$

After substitution of (55) into (53), we find a system of linear equations in the coefficients n_k :

$$n_k(z, v) + \frac{\rho^2}{h^2} \sum_m F_{m, k-m} n_{k-m}(z, v) = N_0 \delta_{0,k}, \tag{56}$$

where

$$F_{m, k-m}(z, v) = \int_0^\infty d\theta f_m(\theta) e^{-i(k-m)\delta\theta}. \tag{57}$$

We shall prove that the system (56) has a unique solution. For the sake of simplicity, we shall limit ourselves to a particular case of (56), i.e., to a system of the form

$$n_k + B_{k, k-1} n_{k-1} + B_{k, k} n_k + B_{k, k+1} n_{k+1} = \delta_{0,k}, \tag{58}$$

but the proof can easily be extended to the general case. Let us suppose that a solution exists, so that each n_k has a finite value which satisfies (58). In particular, n_{-1} will have the value \bar{n}_{-1} . The variables $n_0, n_1, \dots, n_k, \dots$ will then satisfy the linear system

$$\begin{aligned} (1 + B_{0,0})n_0 + B_{0,1}n_1 &= A, \\ B_{1,0}n_0 + (1 + B_{1,1})n_1 + B_{1,2}n_2 &= 0, \\ B_{2,1}n_1 + (1 + B_{2,2})n_2 + B_{2,3}n_3 &= 0, \end{aligned} \tag{59}$$

where

$$A = 1 - B_{0,-1} \bar{n}_{-1}. \tag{60}$$

The infinite determinant of the coefficients of the system (59) is therefore

$$\Delta_0 = \begin{vmatrix} 1 + B_{0,0} & B_{0,1} & 0 & 0 & \dots \\ B_{1,0} & 1 + B_{1,1} & B_{1,2} & 0 & \dots \\ 0 & B_{2,1} & 1 + B_{2,2} & B_{2,3} & \dots \\ \vdots & \vdots & \vdots & \vdots & \dots \end{vmatrix}. \tag{61}$$

One can easily show that the determinant is "normal",¹⁴ in the sense that

$$\sum_{k,m} |B_{k, k-m}| < \infty. \tag{62}$$

In fact, each $B_{k, k-m}$ tends to zero as k^{-2} , when k tends to infinity, while the sum over m ranges from -1 to $+1$, i.e., is a sum of a finite number of terms. These two facts assure us that the sum (62) is convergent. Then the determinant (61) has a finite value; i.e., the succession of determinants

$$\Delta_0^{(1)} = 1 + B_{0,0}, \tag{63}$$

$$\Delta_0^{(2)} = \begin{vmatrix} 1 + B_{0,0} & B_{0,1} \\ B_{1,0} & 1 + B_{1,1} \end{vmatrix}, \tag{64}$$

$$\Delta_0^{(3)} = \begin{vmatrix} 1 + B_{0,0} & B_{0,1} & 0 \\ B_{1,0} & 1 + B_{1,1} & B_{1,2} \\ 0 & B_{2,1} & 1 + B_{2,2} \end{vmatrix} \tag{65}$$

is convergent toward a finite value.

Furthermore, the determinant of n_0 , namely,

$$\Delta_{n_0} = \begin{vmatrix} A & B_{0,1} & 0 & 0 & \dots \\ 0 & 1 + B_{1,1} & B_{1,2} & 0 & \dots \\ 0 & B_{2,1} & 1 + B_{2,2} & B_{2,3} & \dots \\ \vdots & \vdots & \vdots & \vdots & \dots \end{vmatrix}, \tag{66}$$

is also convergent, and it can be shown that its value is

$$\Delta_{n_0} = A \Delta_1, \tag{67}$$

where

$$\Delta_1 = \begin{vmatrix} 1 + B_{1,1} & B_{1,2} & 0 & 0 & \dots \\ B_{2,1} & 1 + B_{2,2} & B_{2,3} & 0 & \dots \\ 0 & B_{3,2} & 1 + B_{3,3} & B_{3,4} & \dots \\ \vdots & \vdots & \vdots & \vdots & \dots \end{vmatrix}. \tag{68}$$

A fundamental theorem in the theory of infinite determinants¹⁴ states that if the determinant of the coefficients is normal [i.e., Eq. (62) holds] and the known terms in the right-hand sides of system (59) are limited, then a unique and limited solution to system (59) exists. In other words, we can assign a finite value to each unknown, and this set of values satisfies (59); furthermore, there is only one set of values which is limited.

Now, if \bar{n}_{-1} is not infinite, then the two requirements above are met, and a limited succession of values for $n_0, n_1, \dots, n_k, \dots$, respectively, is found which satisfies (59).

We now have to determine \bar{n}_{-1} . Before doing this, we find the functional relationship between n_0 and \bar{n}_{-1} . We have, from (60) and (67),

$$n_0 = (\Delta_1 / \Delta_0) (1 - B_{0,-1} \bar{n}_{-1}). \tag{69}$$

Furthermore, after some algebra, one gets from (59)

$$n_1 = -B_{1,0} (\Delta_2 / \Delta_1) n_0, \tag{70}$$

where

$$\Delta_2 = \begin{vmatrix} 1 + B_{2,2} & B_{2,3} & 0 & 0 & \dots \\ B_{3,2} & 1 + B_{3,3} & B_{3,4} & 0 & \dots \\ 0 & B_{4,3} & 1 + B_{4,4} & B_{4,5} & \dots \\ \vdots & \vdots & \vdots & \vdots & \dots \end{vmatrix}. \tag{71}$$

The function $n(t, z, v)$ must be real. Thus, the coefficients n_k must satisfy the relation

$$n_{-k} = n_k^*. \tag{72}$$

This in turn requires that

$$B_{-p, -p-m} = B_{p, p+m}^* \quad (73)$$

According to (72), we choose \bar{n}_{-1} to be the complex conjugate of n_1 . Then, taking the complex conjugate of Eq. (70), and using (73), we get

$$\bar{n}_{-1} = -B_{-1,0}(\Delta_2/\Delta_1)^* n_0.$$

If we substitute this equation into (69) we obtain

$$n_0 = [\Delta_0/\Delta_1 - B_{0,-1} B_{-1,0}(\Delta_2/\Delta_1)^*]^{-1}. \quad (74)$$

The ratio Δ_0/Δ_1 (or more generally the ratio Δ_{k-1}/Δ_k) can be expressed in terms of the ratio Δ_2/Δ_1 (or Δ_{k+1}/Δ_k);

$$\Delta_{k-1}/\Delta_k = 1 + B_{k-1,k-1} - B_{k-1,k} B_{k,k-1} \Delta_{k+1}/\Delta_k, \quad (75)$$

so that

$$n_0 = [1 + B_{0,0} - B_{0,1} B_{1,0} \Delta_2/\Delta_1 - B_{0,-1} B_{-1,0}(\Delta_2/\Delta_1)^*]^{-1}. \quad (76)$$

Using (75) and (73) we can expand n_0 in the continued fraction

$$n_0 = \frac{1}{1 + B_{0,0} - 2 \operatorname{Re} \left(\frac{B_{0,1} B_{1,0}}{1 + B_{1,1} - \frac{B_{1,2} B_{2,1}}{1 + B_{2,2} - \dots}} \right)}. \quad (77)$$

This continued-fraction expansion has been found by Stenholm and Lamb,⁵ and we shall show in Appendix B that our integral-equation formulation of the problem leads to the same result for a single-standing-mode configuration in gas lasers.

Unfortunately, the procedure which led to the continued-fraction expansion (77) cannot be extended directly to the case in which the kernel in Eq. (53) has more than three oscillating terms with frequencies $-\delta$, 0 , and $+\delta$. Thus we shall develop in Sec. V a method which will enable us to treat the multimode case. However, we note that the proof of convergence is valid for any number of oscillating terms in the kernel, because the convergence of the sum (62) is achieved also when m ranges over a larger (but finite) number of terms.

Before concluding this section, we want to emphasize some considerations about the system (59). The determinant of the system (59) is normal, in the sense that the sum (62) is convergent. This fact leads to the existence of a unique solution, in which all coefficients are limited. But other

solutions may be constructed. These solutions, obviously, cannot be limited because of the uniqueness of the limited solution, and must therefore be divergent. Thus, these solutions are non-physical. However, the way other solutions are determined is quite interesting, because it suggests the generalization to multimode operation, which is discussed in Sec. V.

Let us put

$$n_k = n_{-k}^* = 0 \quad \text{for some } k. \quad (78)$$

Then the system (59) splits into two systems, the first of which has k unknowns and k equations:

$$\begin{aligned} (1 + B_{0,0})n_0 + B_{0,1}n_1 &= A, \\ B_{1,0}n_0 + (1 + B_{1,1})n_1 + B_{1,2}n_2 &= 0, \\ &\vdots \\ B_{k-1,k-2}n_{k-2} + (1 + B_{k-1,k-1})n_{k-1} &= 0. \end{aligned} \quad (79)$$

This system can be solved, and we find the corresponding values for n_0, \dots, n_{k-1} . We then introduce in the second system the value obtained for n_{k-1} , and find

$$\begin{aligned} B_{k,k+1}n_{k+1} &= -B_{k,k-1}n_{k-1}, \\ (1 + B_{k+1,k+1})n_{k+1} + B_{k+1,k+2}n_{k+2} &= 0, \\ B_{k+2,k+1}n_{k+1} + (1 + B_{k+2,k+2})n_{k+2} + B_{k+2,k+3}n_{k+3} &= 0, \end{aligned} \quad (80)$$

etc. The determinant of the unknowns n_{k+1}, n_{k+2} , etc. is not normal (the elements $1 + B_{kk}$ are out of the diagonal!). Moreover, it is vanishing,

$$\begin{aligned} \Delta &= \begin{vmatrix} B_{k,k+1} & 0 & 0 & 0 & \cdots \\ 1 + B_{k+1,k+1} & B_{k+1,k+2} & 0 & 0 & \cdots \\ B_{k+2,k+1} & 1 + B_{k+2,k+2} & B_{k+2,k+3} & 0 & \cdots \\ \cdot & \cdot & \cdot & \cdot & \cdots \\ \cdot & \cdot & \cdot & \cdot & \cdots \\ \cdot & \cdot & \cdot & \cdot & \cdots \end{vmatrix} \\ &= B_{k,k+1} B_{k+1,k+2} B_{k+2,k+3} \cdots = 0, \end{aligned} \quad (81)$$

owing to the fact that $B_{p,p+1} \rightarrow 0$ as $p \rightarrow \infty$.

Then the solution of (80) may be unlimited, and in fact it is unlimited. But we note that the value of n_0 obtained from (79) is the same as we would have obtained from (77), just by truncating the continued-fraction expansion at k th order. In fact,

in order to truncate the continued fraction at k th order we must set $B_{k-1,k}$ equal to zero, and the system (59) splits into two systems, the first of which coincides with (79). Thus we can make the following statement: The right value of n_0 can be obtained as the limit of the succession of those values of n_0 belonging to the divergent solutions obtained by setting successively $n_1 = 0, n_2 = 0, \dots, n_k = 0$. It is precisely this statement that can be generalized in order to get the convergent solution in the multimode-operation case. The existence of nonphysical solutions to this problem has been stressed by Feldman and Feld.⁸

V. GENERAL CASE

We now turn to the problem of determining the steady-state solution of the equation

$$n(t, z, v) = N_0 - \frac{\mathcal{P}^2}{\hbar^2} \sum_{m=-N}^{+N} e^{im\delta t} \times \int_0^\infty d\theta f_m(\theta) n(t - \theta, z, v). \tag{82}$$

The solution to (82) may be put in a form similar to (55):

$$n(t, z, v) = \sum_k n_k(z, v) e^{ik\delta t}, \tag{83}$$

and the substitution of (83) into (82) yields the linear system in the unknowns $n_k(z, v)$:

$$\sum_{m=-N}^{+N} \left(\delta_{0,m} + \frac{\mathcal{P}^2}{\hbar^2} F_{m,k-m} \right) n_{k-m} = N_0 \delta_{0,k}, \tag{84}$$

$$k = 0, \pm 1, \pm 2, \dots$$

System (84) is then written in a matrix form; we define the following vectors and matrices:

$$\mathfrak{X}_0 = \begin{pmatrix} n_{-N} \\ n_{-N+1} \\ \vdots \\ n_{-1} \\ n_0 \\ n_1 \\ \vdots \\ n_{N-1} \\ n_N \end{pmatrix}, \tag{85}$$

$$\mathfrak{X}_p = \begin{pmatrix} n_{-pN} \\ n_{-pN+1} \\ \vdots \\ n_{-(p-1)N+1} \\ 0 \\ n_{(p-1)N+1} \\ \vdots \\ n_{pN-1} \\ n_{pN} \end{pmatrix}, \tag{86}$$

$$\mathfrak{J} = \begin{pmatrix} 0 \\ 0 \\ \vdots \\ 0 \\ N_0 \\ 0 \\ \vdots \\ 0 \\ 0 \end{pmatrix}. \tag{87}$$

All vectors (85)–(87) contain $2N+1$ elements. The vector \mathfrak{J} has all elements equal to zero, with the exception of the central one, which equals N_0 ;

$$Z_0 = \begin{pmatrix} 1 + (\mathcal{P}^2/\hbar^2)F_{0,-N} & (\mathcal{P}^2/\hbar^2)F_{-1,-N+1} & \cdots & (\mathcal{P}^2/\hbar^2)F_{-N,0} & \cdots & 0 & 0 \\ (\mathcal{P}^2/\hbar^2)F_{1,-N} & 1 + (\mathcal{P}^2/\hbar^2)F_{0,-N+1} & \cdots & (\mathcal{P}^2/\hbar^2)F_{-N+1,0} & \cdots & 0 & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \vdots \\ (\mathcal{P}^2/\hbar^2)F_{N,-N} & (\mathcal{P}^2/\hbar^2)F_{N-1,-N+1} & \cdots & 1 + (\mathcal{P}^2/\hbar^2)F_{0,0} & \cdots & (\mathcal{P}^2/\hbar^2)F_{-N+1,N-1} & (\mathcal{P}^2/\hbar^2)F_{-N,N} \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & \cdots & (\mathcal{P}^2/\hbar^2)F_{N-1,0} & \cdots & 1 + (\mathcal{P}^2/\hbar^2)F_{0,N-1} & (\mathcal{P}^2/\hbar^2)F_{-1,N} \\ 0 & 0 & \cdots & (\mathcal{P}^2/\hbar^2)F_{N,0} & \cdots & (\mathcal{P}^2/\hbar^2)F_{1,N-1} & 1 + (\mathcal{P}^2/\hbar^2)F_{0,N} \end{pmatrix}, \tag{88}$$

$$U_p = \begin{pmatrix} \mathfrak{G}_p^+ & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \mathfrak{G}_p \end{pmatrix}, \quad (89)$$

$$V_p = \begin{pmatrix} \mathfrak{B}_p^+ & 0 & 0 \\ 0 & p & 0 \\ 0 & 0 & \mathfrak{B}_p \end{pmatrix}, \quad (90)$$

$$W_p = \begin{pmatrix} \mathfrak{C}_p^+ & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \mathfrak{C}_p \end{pmatrix}, \quad (91)$$

where \mathfrak{G}_p , \mathfrak{B}_p , and \mathfrak{C}_p are $N \times N$ matrices and are given by

$$\mathfrak{G}_p = \begin{bmatrix} (\mathcal{P}^2/\hbar^2)F_{-N,(\rho-1)N+1} & 0 & \cdots & 0 \\ (\mathcal{P}^2/\hbar^2)F_{-N+1,(\rho-1)N+1} & (\mathcal{P}^2/\hbar^2)F_{-N,(\rho-1)N+2} & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ (\mathcal{P}^2/\hbar^2)F_{-1,(\rho-1)N+1} & (\mathcal{P}^2/\hbar^2)F_{-2,(\rho-1)N+2} & \cdots & (\mathcal{P}^2/\hbar^2)F_{-N,\rho N} \end{bmatrix}, \quad (92)$$

$$\mathfrak{B}_p = \begin{bmatrix} 1 + (\mathcal{P}^2/\hbar^2)F_{0,(\rho-1)N+1} & (\mathcal{P}^2/\hbar^2)F_{-1,(\rho-1)N+2} & \cdots & (\mathcal{P}^2/\hbar^2)F_{-N+1,\rho N} \\ (\mathcal{P}^2/\hbar^2)F_{1,(\rho-1)N+1} & 1 + (\mathcal{P}^2/\hbar^2)F_{0,(\rho-1)N+2} & \cdots & (\mathcal{P}^2/\hbar^2)F_{-N+2,\rho N} \\ \vdots & \vdots & \vdots & \vdots \\ (\mathcal{P}^2/\hbar^2)F_{N-1,(\rho-1)N+1} & (\mathcal{P}^2/\hbar^2)F_{N-2,(\rho-1)N+2} & \cdots & 1 + (\mathcal{P}^2/\hbar^2)F_{0,\rho N} \end{bmatrix}, \quad (93)$$

$$\mathfrak{C}_p = \begin{bmatrix} (\mathcal{P}^2/\hbar^2)F_{N,(\rho-1)N+1} & (\mathcal{P}^2/\hbar^2)F_{N-1,(\rho-1)N+2} & \cdots & (\mathcal{P}^2/\hbar^2)F_{1,\rho N} \\ 0 & (\mathcal{P}^2/\hbar^2)F_{N,(\rho-1)N+2} & \cdots & (\mathcal{P}^2/\hbar^2)F_{2,\rho N} \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \cdots & (\mathcal{P}^2/\hbar^2)F_{N,\rho N} \end{bmatrix}. \quad (94)$$

The matrices \mathfrak{G}_p^+ , \mathfrak{B}_p^+ , and \mathfrak{C}_p^+ are defined from \mathfrak{G}_p , \mathfrak{B}_p , and \mathfrak{C}_p , respectively, through the relation

$$(\mathfrak{G}_p^+)_{i,k} = (\mathfrak{G}_p)_{N+1-i, N+1-k}^*$$

where the asterisk denotes the complex conjugate.

Finally, the matrix S which projects \mathfrak{X}_0 onto \mathfrak{X}_1 is defined:

$$S\mathfrak{X}_0 = \mathfrak{X}_1. \quad (95)$$

Apart from S , all the matrices defined above are nonsingular.¹⁵ In matrix form, system (84) is then written as follows:

$$Z_0\mathfrak{X}_0 + U_2\mathfrak{X}_2 = \mathcal{J}, \quad (96)$$

$$U_{p+1}\mathfrak{X}_{p+1} + V_p\mathfrak{X}_p + W_{p-1}\mathfrak{X}_{p-1} = 0, \quad (97)$$

$$p = 2, 3, 4, \dots$$

By using (96) and (97) we can therefore express any vector \mathfrak{X}_p in terms of \mathfrak{X}_0 and \mathfrak{X}_1 . For example,

$$\begin{aligned} \mathfrak{X}_3 &= -U_3^{-1}(V_2\mathfrak{X}_2 + W_1\mathfrak{X}_1) \\ &= -U_3^{-1}[V_2U_2^{-1}(\mathcal{J} - Z_0\mathfrak{X}_0) + W_1S\mathfrak{X}_0], \end{aligned} \quad (98)$$

$$\mathfrak{X}_4 = -U_4^{-1}[V_3\mathfrak{X}_3 + W_2V_2U_2^{-1}(\mathcal{J} - Z_0\mathfrak{X}_0)]. \quad (99)$$

Let us consider the succession of vectors $\mathfrak{X}_0^{(k)}$ obtained by setting $\mathfrak{X}_2 = 0$, $\mathfrak{X}_3 = 0$, ..., $\mathfrak{X}_p = 0$, etc., i.e., the succession of vectors $\mathfrak{X}_0^{(k)}$ which satisfy the equations

$$Z_0\mathfrak{X}_0^{(2)} = \mathcal{J}, \quad (100)$$

$$(Z_0 - U_2V_2^{-1}W_1S)\mathfrak{X}_0^{(3)} = \mathcal{J}, \quad (101)$$

$$[Z_0 - U_2(V_2 - U_3V_3^{-1}W_2)W_1S]\mathfrak{X}_0^{(4)} = \mathcal{J}. \quad (102)$$

The vectors $\mathfrak{X}_0^{(k)}$ so obtained will converge, as stated in Sec. IV, to the steady-state solution, the only one which is physically acceptable. Thus the multimode operation may be studied by performing a continued-fraction expansion in a matrix form, i.e., by evaluating

$$Z_1 = U_2 \frac{1}{V_2 - U_3 \frac{1}{V_3 - U_4 \frac{1}{V_4 - \dots} W_3}} W_2 \quad (103)$$

and then solving the system

$$(Z_0 - Z_1 S) \mathfrak{X}_0 = \mathcal{J}. \quad (104)$$

Formula (103) is a generalization of the continued-fraction expansion used in high-intensity laser theory.

The simplest description of the multimode operation is given when we assume $\mathfrak{X}_1 = 0$; i.e., we do not allow the inversion of population density to oscillate at the frequencies $\nu_n - \nu_m$. This description, which is usually called the rate-equation approach, is the zeroth-order solution of our integral-equation formulation, but cannot be obtained directly from (100)–(103). Instead, it may be shown that it is the solution of

$$(1 - S) Z_0 (1 - S) \mathfrak{X}_0 = \mathcal{J}. \quad (105)$$

The lowest-order solution is obtained by setting $U_2 = 0$ or, equivalently, $W_1 = 0$. When this approximation is made, one allows for the interaction of each mode with any other mode, through the coupling with matter. However, the beating term between any pair of modes is assumed to be small enough so that it does not interfere with other terms. The presence of the beating terms causes the inversion population density to have (small) oscillations at their frequencies. This approximation is then justified when $\gamma_{ab} \lesssim \delta$, because in this case each atom can couple no more than two adjacent modes.

We shall show in Sec. VI that when $\gamma_{ab} \ll \delta$ the beating terms are vanishingly small, and a rate-equation approach is fully justified. However, when $\gamma_{ab} \gtrsim \delta$, one must evaluate the complete expansion (103). The successive approximations to Z_1 are found by putting $U_3 = 0$, $U_4 = 0$, and so on. At each step of approximation, one takes into account more oscillating terms in the inversion population density. For example, in the $U_3 = 0$ approximation, one allows the function n to oscillate at frequencies $\delta, 2\delta, \dots, 2N\delta$. The existence of a limited solution to Eq. (82), which has been proved in Sec. IV, assures us that the Fourier spectrum of $n(t, z, \nu)$ found by means of these iterations tends toward a well-defined limit, which is the solution to (82).

All matrices which appear in (103) are nonsingular, and in this section we have used those matrices which allow a compact description of the whole process. For computational purposes, however, it is much better to deal with reduced matrices. We note that every matrix in (103) is of

the form

$$\begin{pmatrix} \mathfrak{M} & & \\ & p & \\ & & \mathfrak{N} \end{pmatrix}, \quad (106)$$

where \mathfrak{M} and \mathfrak{N} are also matrices, so that they are reducible; i.e., their inverse, or the product (or sum) of two of them, gives a matrix of the same form as (106). We have

$$\begin{pmatrix} \mathfrak{M} & & \\ & p & \\ & & \mathfrak{N} \end{pmatrix}^{-1} = \begin{pmatrix} \mathfrak{M}^{-1} & & \\ & p^{-1} & \\ & & \mathfrak{N}^{-1} \end{pmatrix}, \quad (107)$$

$$\begin{pmatrix} \mathfrak{M} & & \\ & p & \\ & & \mathfrak{N} \end{pmatrix} \begin{pmatrix} \mathfrak{M}' & & \\ & p' & \\ & & \mathfrak{N}' \end{pmatrix} = \begin{pmatrix} \mathfrak{M}\mathfrak{M}' & & \\ & pp' & \\ & & \mathfrak{N}\mathfrak{N}' \end{pmatrix}. \quad (108)$$

Thus one can evaluate the simpler expansion

$$Z_1' = \mathfrak{C}_2 \frac{1}{\mathfrak{B}_2 - \mathfrak{C}_3 \frac{1}{\mathfrak{B}_3 - \mathfrak{C}_4 \frac{1}{\mathfrak{B}_4 - \dots} \mathfrak{C}_3}} \mathfrak{C}_1, \quad (109)$$

and then it is possible to get Z_1 ,

$$Z_1 = \begin{pmatrix} (Z_1')^+ & & \\ & q & \\ & & Z_1' \end{pmatrix}, \quad (110)$$

where the operation $(Z_1)^+$ has already been defined for the matrices \mathfrak{C}_p , \mathfrak{B}_p , and \mathfrak{C}_p . The central element q is not relevant, because Z_1 must be multiplied by S on the right-hand side, and that operation leaves the central column equal to zero.

The procedure described above allows one to calculate with any desired accuracy \mathfrak{X}_0 , i.e., the average value n_0 of n and the first $2N$ coefficients of the terms oscillating at $\pm\delta, \dots, \pm N\delta$. If one needs the evaluation of some other coefficient, it turns out to be convenient to adopt the same procedure of the continued fraction, rather than evaluate the corresponding \mathfrak{X}_p vector (which contains the coefficient) from \mathfrak{X}_0 and \mathfrak{X}_1 . This latter calculation does not tell us anything about accuracy.

Suppose we need an evaluation of the coefficient of the term oscillating as $e^{iqa\delta t}$. Multiplying both sides of (82) by $e^{iqa\delta t}$, we get the integral equation for the function

$$n^{(a)}(t, z, \nu) = e^{iqa\delta t} n(t, z, \nu), \quad (111)$$

i.e.,

$$n^{(a)}(t, z, \nu) = N_0 e^{i q \delta t} - \frac{\mathcal{P}^2}{\hbar^2} \sum_{m=-N}^{+N} e^{i m \delta t} \int_0^\infty d\theta [f_m(\theta) e^{i q \delta t} n^{(a)}(t - \theta, z, \nu)]. \quad (112)$$

This equation can be treated in the same way as (82), with the only difference being that the source term is an oscillating function of time. This in turn modifies Eqs. (96) and (97), in the sense that the source term will appear in Eq. (97) for some p . Obviously, one has to take into account the source term, so that one cannot truncate the expansion which is obtained from (103) before that order p . For example, the lowest-order approximation to n_{3N+1} will be achieved by putting $\mathcal{K}_5 = 0$ (which appears first in the $p = 4$ equation, where the right-hand side is different from zero).

VI. APPLICATION: MULTIMODE LASER OPERATION IN AN ACTIVE MEDIUM WITH UNMOVING ATOMS

Here we shall apply our formalism to the case of laser operation in a medium which has active atoms (or molecules) fixed in some lattice sites; the line of gain may equally well be nonhomogeneous, because local perturbations (such as elec-

tric field, or stress in the crystal lattice) may change the atomic energy levels, so that two different atoms in two different sites may have different transition frequencies. The macroscopic density matrix $\rho(z, t, \omega)$ will then describe the temporal behavior of those atoms placed in a small volume around z , which have the same transition frequency ω . The source term in the integral equation, N_0 , will also be ω dependent; we assume that the ω dependence of the density matrix is not changed by the laser electric field. Thus the macroscopic polarization (38) will be given by a sum (integral) over the set of ω values which the atoms of the medium may assume:

$$P(z, t) = \int d\omega \mathcal{P}[\rho_{ab}(z, t, \omega) + \rho_{ba}(z, t, \omega)]. \quad (113)$$

For a standing-wave configuration of the electric field in the cavity, we must use expression (A6) for the kernel in the integral equation, with $\nu = 0$:

$$K(t, \theta) = \sum_{n, n'} \sum_{\alpha} \frac{\frac{1}{2} E_n E_{n'} \sin k_n z \sin k_{n'} z \exp[i(\phi_n - \phi_{n'})]}{i(\nu_n - \omega) + \gamma_\alpha - \gamma_{ab}} \times \exp[-i(\nu_{n'} - \nu_n)t] \{ \exp[-(\gamma_{ab} + i\omega - i\nu_{n'})\theta] - \exp(-\gamma_\alpha \theta) \} + \text{c.c.} \quad (114)$$

The kernel (114) has the form (A7) but, owing to the stationary-atoms condition, the complex constants Γ_p which appear there are z independent. We can assume that the modes have equal frequency separation, i.e.,

$$\nu_{n+1} - \nu_n = \delta. \quad (115)$$

One can readily verify that the integral equation (35) assumes the form (53). The sum over m runs from $-M+1$ to $M-1$, where M is the number of modes operating in the cavity, so that we put

$$N = M - 1. \quad (116)$$

When there is one mode in the cavity, the kernel (114) becomes independent of t . The situation is quite similar to that discussed in Sec. III: the inversion population density is then time independent, and its value may be found directly,

$$n(z, \omega) = N_0 / \Delta, \quad (117)$$

with

$$\Delta = 1 + \frac{\mathcal{P}^2}{\hbar^2} \frac{E^2 \sin^2 kz}{2} \frac{\gamma_{ab}(\gamma_a + \gamma_b)}{\gamma_a \gamma_b} \frac{1}{\gamma_{ab}^2 + (\omega - \nu)^2}.$$

Note that the inversion population density in (117) is dependent on z , just because the standing-wave configuration of the electromagnetic field has a spatial modulation of the time-averaged intensity, which in turn determines a spatial modulation of the inversion density.

When two modes oscillate in the cavity, the kernel has three terms dependent on t , with frequencies $-\delta$, 0 , and $+\delta$. The structure of the kernel is such that each Fourier component of n is coupled with its two nearest neighbors, i.e., with the components whose frequencies differ by $\pm\delta$.

The linear system for the Fourier coefficients has the form (58), for which the solution of n_0 has been obtained as a continued-fraction expansion. Details of these calculations will be given elsewhere.

In the general case of M -mode oscillation, we have the prescription for the evaluation of the matrix elements $F_{m, k-m}$. We have

$$f_m(\theta) = \sum_{\substack{n, n' \\ n - n' = m}} \sum_{\alpha} \left[\frac{1}{4} E_n E_{n'} \sin k_n z (\sin k_{n'} z) e^{i(\phi_n - \phi_{n'})} \right] \\ \times \left(\frac{1}{i(\nu_n - \omega) + \gamma_{\alpha} - \gamma_{ab}} (e^{-(\gamma_{ab} + i\omega - i\nu_{n'})\theta} - e^{-\gamma_{\alpha}\theta}) \right. \\ \left. + \frac{1}{-i(\nu_{n'} - \omega) + \gamma_{\alpha} - \gamma_{ab}} (e^{-(\gamma_{ab} - i\omega + i\nu_n)\theta} - e^{-\gamma_{\alpha}\theta}) \right), \quad (118)$$

$$F_{m,k-m} = \sum_{\substack{n, n' \\ n - n' = m}} \sum_{\alpha} \left[\frac{1}{4} E_n E_{n'} \sin k_n z (\sin k_{n'} z) e^{i(\phi_n - \phi_{n'})} \right] \\ \times \left[\frac{1}{\gamma_{ab} - i\omega + i\nu_{n'} + ik\delta} \frac{1}{\gamma_{\alpha} + i(k-m)\delta} \left(1 - \frac{im\delta}{-i(\nu_{n'} - \omega) + \gamma_{\alpha} - \gamma_{ab}} \right) \right. \\ \left. + \frac{1}{\gamma_{ab} + i\omega - i\nu_n + ik\delta} \frac{1}{\gamma_{\alpha} + i(k-m)\delta} \left(1 - \frac{im\delta}{i(\nu_n - \omega) + \gamma_{\alpha} - \gamma_{ab}} \right) \right]. \quad (119)$$

The behavior of $F_{m,k-m}$, when $k \rightarrow \infty$, is readily found from (119). The Lorentzian factors may be combined to give

$$F_{m,k-m} \simeq \sum_{\substack{n, n' \\ n - n' = m}} \sum_{\alpha} \left[\frac{1}{4} E_n E_{n'} \sin k_n z (\sin k_{n'} z) e^{i(\phi_n - \phi_{n'})} \right] \\ \times \left[\frac{\gamma_{\alpha} - \gamma_{ab} - i(\nu_{n'} - \omega) - im\delta}{\gamma_{\alpha} - \gamma_{ab} - i(\nu_{n'} - \omega)} + \frac{\gamma_{\alpha} - \gamma_{ab} + i(\nu_n - \omega) - im\delta}{\gamma_{\alpha} - \gamma_{ab} + i(\nu_n - \omega)} \right] \left(-\frac{1}{k^2 \delta^2} \right) (k \rightarrow \infty), \quad (120)$$

which shows the $1/k^2$ dependence when $k \rightarrow \infty$ (see Sec. IV).

From (119) we can explain a lot of features which appear in multimode operation. Let us consider the case $\gamma_{\alpha} \ll \delta$ ($\alpha = a, b$). This means that the lifetimes of the levels are long compared with the period $1/\delta$ of oscillation of the beating term of two adjacent modes. When this condition is satisfied, one would expect that the inversion population density does not oscillate, because all beating terms vanish due to interference in a level lifetime. But this is not the case. In fact, from (119) it is apparent that the resonance factor

$$1/[\gamma_{\alpha} + i(k-m)\delta]$$

is appreciably different from zero when $m = k$. Then in the system (56) all $F_{m,k-m}$ may be put equal to zero, with the exception of $F_{m,0}$. This means that the Fourier coefficients n_k [$k = 0, \pm 1, \dots, \pm(M-1)$] are coupled with n_0 , and the others are zero. The average value of the inversion population density is therefore given by

$$n_0 [1 + (\mathcal{P}^2/\hbar^2) F_{0,0}] = N_0, \quad (121)$$

which reads

$$n_0 = N_0/\Delta', \quad (122)$$

$$\Delta' = 1 + \frac{\mathcal{P}^2}{\hbar^2} \sum_n \frac{1}{2} E_n^2 \sin^2 k_n z \\ \times \frac{\gamma_{ab}(\gamma_a + \gamma_b)}{\gamma_a \gamma_b} \frac{1}{(\omega - \nu_n)^2 + \gamma_{ab}^2},$$

while the other coefficients n_k are given by

$$n_k = -(\mathcal{P}^2/\hbar^2) F_{k,0} n_0. \quad (123)$$

If we compare (122) with (117), we see that the average population density is not affected by the beating terms, and is simply saturated by all the modes which act independently. But Eq. (123) indicates that some oscillations arise in the population density. These oscillations are due to the fact that the beating terms influence the polarization of the medium, whose lifetime γ_{ab}^{-1} may be comparable with or shorter than the period of oscillation $1/\delta$. The polarization, in turn, influences the population density, and induces oscillations with frequencies $\nu_n - \nu_m$. However, the population-density oscillations do not induce further oscillations in the polarization, because of its long lifetime, and the chain of interactions stops here.

The situation described above, $\gamma_{\alpha} \ll \delta$ ($\alpha = a, b$), is met in several solid-state laser devices, such as a ruby laser operating at 6943 Å. The lifetime of the excited level of ruby is much longer (10^{-3} sec) than the period of oscillation $1/\delta$ (which is usually of the order of 10^{-8} sec), so that the condition $\gamma_{\alpha} \ll \delta$ is well satisfied.¹⁶ On the other hand, the lifetime γ_{ab}^{-1} of the polarization is shorter than $1/\delta$ (γ_{ab} is the homogeneous bandwidth of the line of gain, and it is of the order of 10^9 sec⁻¹, so that $\gamma_{ab}^{-1} \sim 10^{-9}$ sec). Then we can treat the multimode operation in a ruby laser by assuming that

the average population density n_0 is given by (122), and allowing for oscillations in $\mathbf{n}(t, z, \omega)$. Therefore we have

$$n(z, t, \omega) = n_0(z, \omega) \left(1 - \frac{\mathcal{P}^2}{\hbar^2} \sum_{\substack{k=-M+1 \\ k \neq 0}}^{M-1} F_{k,0} e^{ik\delta t} \right). \quad (124)$$

The polarization $P(t, z, \omega)$ can be evaluated by introducing (124) into (31) and performing the integral.

Obviously when the lifetime of the polarization γ_{ab}^{-1} is also much longer than the period of oscillation $1/\delta$, i.e., when $\gamma_{ab} \ll \delta$, oscillations in the polarization must disappear, and therefore also the population density must have no oscillation. In this case the resonance factor in Eq. (119),

$$1/[\gamma_{ab} \pm i(\omega - \nu_n) + ik\delta],$$

is appreciably different from zero when $k=0$. The only $F_{m,k-m}$ term which survives is therefore $F_{0,0}$. The average population density is given again by (122), but now $n_k = 0$ ($k \neq 0$).

The latter case is usually referred to as the rate-equation approximation. Here the inversion population density has a stationary value n_0 , and all calculations can be done directly, starting from the equations of motion (30) and (31), by putting there $\rho_{aa} - \rho_{bb} = \text{const}$. In our general formalism, the rate-equation approximation is given by Eq. (105).

The rate-equation approximation is also suitable for treating the free-running operation. In the free-running operation, the phase factor $e^{-i\phi_n}$ of the n th mode is oscillating randomly in time owing to the interaction among the modes. One can therefore substitute the average value $\langle e^{i(\phi_n' - \phi_n)} \rangle$ in place of $e^{i(\phi_n' - \phi_n)}$ in Eq. (119), and assume that it is zero, except for $n=n'$. Then the interference terms among the modes vanish, and one has

$$F_{m,k-m} = F_{0,k} \delta_{m,0}. \quad (125)$$

In this case, each Fourier coefficient n_k is coupled only with itself [see Eq. (56)], but the source term N_0 is present only in the equation for n_0 , so that only n_0 is different from zero, while the other coefficients n_k are zero. The average population density is given by (122) again, although what causes the oscillating terms to give no contribution to n_0 is different in the two cases.

In the general case one must use the matrix continued-fraction expansion of Sec. V in order to find the population density. However, one can reduce the order of the matrices involved if the lifetimes of the levels are sufficiently long, so that

$$\gamma_\alpha < P\delta \quad (126)$$

for some P . Then one can assume

$$F_{m,k-m} \simeq 0 \quad (127)$$

for $|m| > P$. The sum in Eq. (56) is then limited to values of m which run from $-P$ to $+P$. In most cases, P is equal to several units (say 3 or 4) and the multimode analysis turns out to be quite tractable with the general formalism we have developed. We shall give detailed calculations for the multimode operation in a forthcoming paper.

One other limiting case, i.e., the close-coupling approximation (CCA), will be described here. If we are in a situation where

$$M\delta \ll \gamma_{ab}, \quad (128)$$

$$M\delta \ll \gamma_\alpha, \quad (129)$$

i.e., if the modes are limited in a small fraction of the total bandwidth, we can make a Markoffian approximation in the integral equation; i.e., we can substitute $\mathbf{n}(t, z, \omega)$ for $\mathbf{n}(t - \theta, z, \omega)$ in the integrand of (82). This is justified by the fact that the memory time of the population inversion is much smaller than its period of oscillation; i.e., the kernel decays to zero in a time in which n has changed very little.

The CCA gives the expression for $\mathbf{n}(t, z, \nu)$:

$$n(t, z, \nu) = \frac{N_0}{1 + (\mathcal{P}^2/\hbar^2) \sum_m F_{m,0} e^{im\delta t}}. \quad (130)$$

This approximation turns out to be very useful in a situation where the modes merge in a quasi-continuum distribution.

VII. CONCLUSIONS

A method for treating the interaction between a two-level system and a multimode e.m. field has been described, and is now summarized, with some of its advantages pointed out.

(i) It unifies the semiclassical theories, because one can readily obtain from it both the perturbation expansion in a power series of the e.m. field (Sec. III) and the continued-fraction expansion (Sec. IV).

(ii) The continued-fraction expansion in matrix form, which has been derived from it, makes it possible to treat the multimode operation without any upper bound to the e.m. field. This solution turns out to be always convergent.

(iii) Some particular cases can be treated (Sec. VI) without carrying out the matrix expansion. For solid-state laser devices, such as ruby lasers, with a long lifetime of the excited level, I have shown how to construct the solution to the multimode case in a very simple way.

(iv) The method itself is quite general, and can be applied to many physical problems where the all-order interaction between matter and the e.m. field must be taken into account.

Only a few examples have been discussed, but I think that the ability of the method of covering a large class of phenomena has been sufficiently stressed. Numerical calculations will be reported elsewhere.¹⁷

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APPENDIX A

In this appendix we shall find the kernel [Eq. (36)] which enters into the integral equation for $n(t, z, v)$ [Eq. (35)]. Two typical situations are taken into account. In the first one, the field is in a superposition of running waves (ring-laser operation). In the second one, the field is made up of standing modes (Fabry-Perot laser cavities).

The running-wave configuration has spatial mode functions $U_n(z)$ given by Eq. (17); the kernel is then given by

$$K_{RW}(t, t'') = \frac{1}{4} \int_{t''}^t dt' \sum_{n, n'} E_n E_{n'} \{ \exp[-i(\nu_n t' + \phi_n - k_n z')] \exp[-i(\nu_{n'} t'' + \phi_{n'} - k_{n'} z)] + \exp[i(\nu_n t' + \phi_n - k_n z')] \exp[-i(\nu_{n'} t'' + \phi_{n'} - k_{n'} z)] + c.c. \} \times (e^{-\gamma_a(t-t'')} + e^{-\gamma_b(t-t'')}) \{ \exp[-(\gamma_{ab} + i\omega)(t' - t'')] + \exp[-(\gamma_{ab} - i\omega)(t' - t'')] \}, \tag{A1}$$

where the index RW stands for running wave.

In deriving (A1), we have put $z' = z - v(t - t')$ and $z'' = z' - v(t' - t'') = z - v(t - t'')$. The field amplitude labeled by n' is then independent of t' , and the kernel may be simplified:

$$K_{RW}(t, t'') = E(z'', t'') \frac{1}{2} \sum_n \int_{t''}^t dt' E_n \{ \exp[-i(\nu_n t' + \phi_n - k_n z)] + c.c. \} (e^{-\gamma_a(t-t'')} + e^{-\gamma_b(t-t'')}) \times \{ \exp[-(\gamma_{ab} + i\omega)(t' - t'')] + \exp[-(\gamma_{ab} - i\omega)(t' - t'')] \}. \tag{A2}$$

The integral in (A2) can be easily performed. The result is

$$K_{RW}(t, t'') = E(z'', t'') \left(\sum_{\alpha=a, b} \frac{\frac{1}{2} E_n e^{-i\phi_n}}{-i(\nu_n - k_n v) + \gamma_\alpha - \gamma_{ab} - i\omega} \{ \exp[-i(\nu_n t - k_n z) - (\gamma_{ab} + i\omega)(t - t'')] - \exp[-i(\nu_n t'' - k_n z'') - \gamma_\alpha(t - t'')] \} + \sum_{\alpha=a, b} \frac{\frac{1}{2} E_n e^{i\phi_n}}{i(\nu_n - k_n v) + \gamma_\alpha - \gamma_{ab} - i\omega} \{ \exp[i(\nu_n t - k_n z) - (\gamma_{ab} + i\omega)(t - t'')] - \exp[i(\nu_n t'' - k_n z'') - \gamma_\alpha(t - t'')] \} \right) + c.c. \tag{A3}$$

The expression (A3) can be further simplified if one neglects the counterrotating waves in the polarization. In this approximation, which is usually referred to as the rotating-wave approximation (RWA), one assumes that the density matrix element ρ_{ab} oscillates at the optical frequency

$$\rho_{ab} = \rho_0 e^{-i\omega t}, \tag{A4}$$

and ρ_0 is a slowly varying function of time. In the RWA one can evaluate the integral equation (35) and its kernel [Eq. (36)] by coupling ρ_{ab} in the equations of motion (27)–(29) with that component $E^{(-)}$ of $E(z, t)$ which oscillates as $e^{i\omega t}$, and ρ_{ba} with the other part of $E(z, t)$. This in turn is equivalent to neglecting terms which oscillate at twice the optical frequency, both in t and t'' in Eq. (A3).

In the RWA, the kernel is therefore given by

$$\begin{aligned}
K_{\text{RW}}(t, t'') = & \sum_n \sum_{n'} \sum_{\alpha=a,b} \frac{1}{4} \frac{E_n E_{n'} e^{i(\phi_n - \phi_{n'})}}{i(\nu_n - k_n v - \omega) + \gamma_\alpha - \gamma_{ab}} \\
& \times \{ \exp[-i(\nu_n t'' - k_n z'')] \exp[i(\nu_n t - k_n z)] \exp[-(\gamma_{ab} + i\omega)(t - t'')] \} \\
& - \{ \exp[i(\nu_n t'' - k_n z'')] \exp[-i(\nu_n t'' - k_n z'')] \exp[-\gamma_\alpha(t - t'')] \} + \text{c.c.} \quad (\text{A5})
\end{aligned}$$

We can derive similar formulas in a standing-wave configuration of the e.m. field. Since each standing mode is made up of two running waves, with the same frequency ν_n and with opposite propagation vector K_n , it is possible to obtain the kernel $K(t, t'')$ by simply extending the summation in (A3) or in (A5) over a parameter μ which can assume the values +1 and -1. An extra factor $-\frac{1}{2}i\mu$ for each field will then provide the right dependence of the standing mode on z [Eq. (18)]. For example, we give the expression for the kernel in a standing-wave (SW) configuration, in the RWA:

$$\begin{aligned}
K_{\text{SW}}(t, t'') = & \sum_n \sum_{n'} \sum_{\alpha=a,b} \sum_{\mu=\pm 1} \sum_{\mu'=\pm 1} \frac{1}{4} \frac{-\frac{1}{2}i\mu'(\frac{1}{2}i\mu)E_n E_{n'} e^{i(\phi_n - \phi_{n'})}}{i(\nu_n - \mu k_n v - \omega) + \gamma_\alpha - \gamma_{ab}} \\
& \times \{ \exp[-i(\nu_n t'' - \mu' k_n z'')] \exp[i(\nu_n t - \mu k_n z)] \exp[-(\gamma_{ab} + i\omega)(t - t'')] \} \\
& - \exp[+i(\nu_n t'' - \mu k_n z'')] \exp[-i(\nu_n t'' - \mu' k_n z'')] \exp[-\gamma_\alpha(t - t'')] + \text{c.c.} \quad (\text{A6})
\end{aligned}$$

We note that the damping factors enter expressions (A5) or (A6) only through the exponentials $e^{-\gamma(t-t'')}$ (where γ may be γ_{ab} , γ_a , or γ_b). Then one can make the substitution

$$t - t'' = \theta$$

and the kernel turns out to be a function of the two variables t and θ ,

$$K(t, \theta) = \sum_p A_p(z, v) e^{i\Omega_p t} e^{\Gamma_p \theta}, \quad (\text{A7})$$

where the A_p are complex coefficients which depend on v and z , the Ω_p are real frequencies given by

$$\Omega_p = \nu_n - \nu_{n'}, \quad (\text{A8})$$

and the Γ_p are complex constants whose real parts are $-\gamma_{ab}$, $-\gamma_a$, or $-\gamma_b$.

So far, we have derived the integral equation in its general form, which we shall write here:

$$\begin{aligned}
n(t, z, v) = & N_0 - \frac{\mathcal{P}^2}{\hbar^2} \sum_p A_p(z, v) e^{i\Omega_p t} \\
& \times \int_0^\infty d\theta e^{\Gamma_p \theta} n(t - \theta, z - v\theta, v).
\end{aligned}$$

In the integrand, the population density n depends on θ through t and z . This is a consequence of the condition (25) which we have imposed on z_0 and t_0 . Then the contribution to n at time t and position z comes from the population density of those

atoms which were at the position $z - v\theta$ at the time $t - \theta$. Owing to the atomic motion, a variation of the z coordinate turns out to be equivalent to a shift of the frequencies of the e.m. field. This fact may be used to find an integral equation where the θ dependence of n is displayed only through $t - \theta$. Let us introduce the coordinate

$$\zeta = z(0),$$

where the atoms we are considering were at time $t = 0$. The integral equation for the population density of these atoms is therefore found by substituting $\zeta + vt$ in place of z . This substitution yields

$$\begin{aligned}
n(t, \zeta + vt, v) \\
= & N_0 - \frac{\mathcal{P}^2}{\hbar^2} \sum_p A_p(\zeta + vt, v) e^{i\Omega_p t} \\
& \times \int_0^\infty d\theta e^{\Gamma_p \theta} n(t - \theta, \zeta + v(t - \theta), v),
\end{aligned}$$

which may be written

$$\begin{aligned}
n(t, \zeta, v) = & N_0 - \frac{\mathcal{P}^2}{\hbar^2} \sum_p A_p(\zeta, v) e^{i\Omega'_p t} \\
& \times \int_0^\infty d\theta e^{\Gamma_p \theta} n(t - \theta, \zeta, v). \quad (\text{A9})
\end{aligned}$$

The Ω'_p are the new frequencies, shifted by the Doppler effect. We note that ζ enters (A9) as a parameter on which the kernel depends. The pop-

ulation density has a unique functional dependence on t .

APPENDIX B

Here we shall derive the continued-fraction expansion for n_0 in the single-mode standing-wave

$$K_{\text{sw}}(t, t'') = \sum_{\alpha} \sum_{\mu} \sum_{\mu'} \frac{E^2}{16} \mu \mu' \frac{1}{i(\nu - \mu k v - \omega) + \gamma_{\alpha} - \gamma_{ab}} e^{i(\mu' - \mu)kz} \times \{ \exp[-(\gamma_{ab} + i\omega - i\nu + i\mu'kv)(t - t'')] - \exp[-(\gamma_{\alpha} + i\mu'kv - i\mu kv)(t - t'')] \} + \text{c.c.}, \quad (\text{B1})$$

where we have dropped the index n in ν_n and k_n .

The kernel depends only on the difference $t - t'' = \theta$, so that the integral equation can be written in the form

$$n(t, z, v) = N_0 - \sum_{p=-1}^1 e^{2ipkz} \times \int_0^{\infty} f_p(\theta) n(t - \theta, z - v\theta, v) d\theta, \quad (\text{B2})$$

where we have written explicitly the z dependence of the kernel.

As noted in Appendix A, this integral equation could be transformed into a form such that n depends on θ only through $t - \theta$. But for a single mode it is much better to use (B2). In fact, the kernel in (B2) is independent of t , so that we are allowed to assume a steady-state solution for n independent of t . The spatial modulation of the kernel, in this case, couples the spatial Fourier coefficients $n(z, v)$, whose expansion is now

$$n(z, v) = \sum n_p(v) e^{ipkvz}. \quad (\text{B3})$$

Note that the index p in (B3) runs over even numbers, because the odd terms form a homogeneous system and may be put equal to zero. The inhomogeneous (source) term N_0 appears only in the system for the set n_p , with p even. The expressions for $f_p(\theta)$ can be readily deduced from (B1).

configuration. The results presented here are identical with those obtained by Stenholm and Lamb⁵ if their continued fraction is "contracted" in a more rapidly convergent expansion.

In the RWA, the kernel of the integral equation for one standing mode is given by (see Appendix A)

If we substitute (B3) into (B2), we get the following system of equations for the coefficients n_p :

$$n_p = N_0 \delta_{0,p} - \frac{\mathcal{G}^2 E^2}{4\hbar^2} \frac{\mathcal{L}_p}{\mathcal{L}_p^2 - \gamma^2} \times (A_p n_{p+2} + B_p n_p + C_p n_{p-2}), \quad (\text{B4})$$

where

$$\mathcal{L}_p = ipkv + \gamma_{ab}, \quad (\text{B5})$$

$$A_p = -\mathcal{L}_{p+1} / (\mathcal{L}_{p+1}^2 + \Omega^2), \quad (\text{B6})$$

$$C_p = -\mathcal{L}_{p-1} / (\mathcal{L}_{p-1}^2 + \Omega^2), \quad (\text{B7})$$

$$B_p = -(A_p + C_p), \quad (\text{B8})$$

$$\Omega = \omega - \nu, \quad (\text{B9})$$

$$\gamma = \frac{1}{2}(\gamma_a - \gamma_b). \quad (\text{B10})$$

In deriving (B4) we have made use of the assumptions of Ref. 5, i.e., a pure radiative decay for the off-diagonal matrix elements, which allows us to write

$$\gamma_{ab} = \frac{1}{2}(\gamma_a + \gamma_b). \quad (\text{B11})$$

The system (B4) admits a solution in a continued-fraction expansion, as we derived in Sec. IV.

After some calculations, one gets

$$n_0 = \frac{N_0}{1 + (\gamma_{ab}/\gamma_a\gamma_b)[B_0 + (A_0\Phi + \text{c.c.})]}, \quad (\text{B12})$$

where

$$\Phi = \frac{n_2}{n_0} = \frac{-\mathcal{L}_2 C_2}{\mathcal{L}_2^2 - \gamma^2 + \mathcal{L}_2 B_2 + \mathcal{L}_2 A_2} \frac{(-\mathcal{L}_4 C_4)}{\mathcal{L}_4^2 - \gamma^2 + \mathcal{L}_4 B_4 + \mathcal{L}_4 A_4} \frac{(-\mathcal{L}_6 C_6)}{\dots}. \quad (\text{B13})$$

Expression (B12) must be compared with Eq. (76) of Ref. 5, which reads

$$n_0 = N_0 (1 + 2 \text{Re}\Psi)^{-1}, \quad (\text{B14})$$

where

$$\Psi = \frac{ID(0)D(1)}{1 + \frac{ID(1)D(2)}{1 + \frac{ID(2)D(3)}{1 + \dots}}}. \quad (\text{B15})$$

The adimensional field intensity I is given by

$$I = (\mathcal{O}^2 E^2 / \hbar^2) (1/2\gamma_a\gamma_b), \quad (\text{B16})$$

$$D(p) = \frac{1}{2} (\frac{1}{2}\gamma_a\gamma_b)^{1/2} [(i p k v + \gamma_a)^{-1} + (i p k v + \gamma_b)^{-1}], \quad p \text{ even}, \quad (\text{B17})$$

$$D(p) = \frac{1}{2} (\frac{1}{2}\gamma_a\gamma_b)^{1/2} \{ [i p k v - i(\omega - \nu) + \gamma_{ab}]^{-1} + [i p k v + i(\omega - \nu) + \gamma_{ab}]^{-1} \}, \quad p \text{ odd}. \quad (\text{B18})$$

The two expressions (B12) and (B14) are identical, as one can show by using the identity (a contraction of the continued fraction)

$$\frac{ID(0)D(1)}{1 + \frac{ID(1)D(2)}{1 + \frac{ID(2)D(3)}{1 + \dots}}} = ID(0)D(1) - \frac{I^2D(0)D^2(1)D(2)}{1 + ID(1)D(2) + ID(2)D(3) - \frac{I^2D(2)D^2(3)D(4)}{1 + ID(3)D(4) + ID(4)D(5) - \frac{I^2D(4)D^2(5)D(6)}{1 + \dots}}}, \quad (\text{B19})$$

and noting that the $D(p)$'s can be expressed in terms of \mathcal{L}_p 's:

$$D(p) = (\frac{1}{2}\gamma_a\gamma_b)^{1/2} \mathcal{L}_p / (\mathcal{L}_p^2 + \Omega^2), \quad p \text{ odd}, \quad (\text{B20})$$

$$D(p) = (\frac{1}{2}\gamma_a\gamma_b)^{1/2} \mathcal{L}_p / (\mathcal{L}_p^2 - \gamma^2), \quad p \text{ even}. \quad (\text{B21})$$

It is more convenient to express the continued fraction in the form (B13) rather than (B15), because one can show its convergence on the basis of the discussion in Sec. IV. But, in fact, the two expressions coincide.

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¹⁶When the atomic excited level has a long lifetime, as in the ruby-laser case, it is not possible to assume that the electric field amplitude changes little in an atomic lifetime; then, in treating transient regimes, one must deal with the exact integral equation, Eqs. (35) and (36). In the steady-state regime, however, the electric field amplitudes do not change with time, and one is allowed to use Eq. (114) for the kernel.

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