

Higher-Order Perturbation Theory in Gaseous Lasers*

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The Lamb theory of the optical maser is extended to fifth and higher orders in the perturbation by the time-dependent iteration method, and by a rate-constant approach obtained by removing the time dependence in the Hamiltonian with a unitary transformation. The third- and fifth-order Fourier projections of the atomic polarization are integrated exactly over the atomic velocity distribution, assumed Maxwellian, and the results are then valid for any ratio of natural linewidth, or cavity detuning, to the Doppler width. Dominant fifth-order terms occur whose variation with cavity detuning depends only on the natural linewidth of the transition. These produce an increase in laser intensity versus cavity detuning, including a reduction of the dip phenomena, and are effective at low levels of laser excitation. In addition, third-order and numerous fifth-order terms occur which involve sharper resonances near line center because of Doppler interference effects. Such terms involve the individual decay constants of the states in a complicated way. Owing to cancellation effects, the totality of such terms is in general small compared with the usual saturation term and the dominant fifth-order contributions. This considerably simplifies the deduction of higher-order perturbation terms. Collision processes would also reduce their effect as a result of their higher-order atomic response functions, but the theory is reasonably consistent with present experimental characteristics even without this possibility. The rate-constant approach facilitates the deduction of the various higher-order perturbation results and would be of even greater utility in discussing the Zeeman laser, particularly for axial magnetic fields and complicated transitions.

1. INTRODUCTION

A THIRD-ORDER perturbation treatment for the atomic polarization generally suffices to determine the steady state of the laser oscillator at low levels of intensity. Such an analysis, however, gives no indication of the level of laser intensity, or excitation, for which the results, such as steady-state intensity and frequency as functions of cavity tuning, are valid. It is thus desirable to extend the analysis to at least fifth order in the perturbation, and to assess the magnitude of the changes which then occur in the steady-state conditions of the laser. This is particularly important for further detailed applications of the theory, and for work in which comparisons between theory and experiment are made by the empirical variation of the atomic parameters, so as to obtain better agreement with the experimental data.

The higher-order perturbation terms are also pertinent to discussions involving mode interactions and polarization changes in Zeeman lasers,¹⁻³ particularly under conditions of neutral coupling between say circularly polarized oscillations in an axial magnetic field.^{2,4} For such noncoarse equilibrium states,⁵ in addition to changes produced by other small perturbations such as in reflector characteristics, the higher-order terms can play a significant role in changing the characteristics of the phase paths of the nonlinear equations in the phase plane. However, while our results may be extended to such cases, we shall not

discuss such applications to the Zeeman laser in the present account.

Some results on the effect of higher-order perturbations on the laser have been given by Uehara and Shimoda.⁶ These were mainly concerned with the Doppler limit, although some higher-order terms in the ratios of linewidth and cavity detuning to the Doppler parameter were considered. Their results showed that the Lamb dip⁷ is affected by the fifth-order terms for threshold parameters as low as 1.10, and that the steady-state intensity deduced from the fifth-order approximation was greater in general than that deduced from the third-order results. A dependence of the steady-state conditions on the ratio γ_a/γ_b of the decay constants of the atomic states, as well as on the usual mean value γ_{ab} , was also noted. It is apparent that such results will affect the values of decay constants, or other atomic parameters which are deduced from the third-order results, and also that some further assessment of the relative magnitude of the various fifth-order terms is necessary, particularly for those which exhibit a dependence on the individual decay constants of the atomic states.

We shall thus be concerned with a more exact deduction of the fifth-order perturbation terms in a two-level atomic transition. First the results will be derived by the time-dependent iteration method following Lamb's procedure, together with the exact integration over the atomic velocity distribution (assumed Maxwellian) of the various terms which occur in the spatial Fourier projection of the atomic polarization onto the single cavity mode of oscillation considered. These deductions will then be valid for any ratio of

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¹ R. L. Fork and M. Sargent, III, *Phys. Rev.* **139**, A617 (1965).

² C. V. Heer and R. D. Graft, *Phys. Rev.* **140**, A1088 (1965).

³ W. Culshaw and J. Kannelaud, *Phys. Rev.* **145**, 257 (1966).

⁴ W. Culshaw and J. Kannelaud, *Phys. Rev.* **156**, A308 (1967).

⁵ A. A. Andronov, A. A. Vitt, and S. E. Khaikin, *Theory of Oscillators* (Pergamon Press Inc., New York, 1966), pp. 374-404.

⁶ K. Uehara and K. Shimoda, *Japan. J. Appl. Phys.* **4**, 921 (1965).

⁷ W. E. Lamb, Jr., *Phys. Rev.* **134**, A1429 (1964).

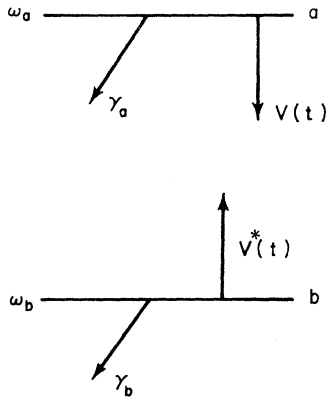


FIG. 1. Two-level laser system and associated parameters.

Doppler width to natural linewidth or cavity detuning. Next we develop what might be termed a rate constant approach to the problem by removing the more rapid time dependence of the optical perturbation by a unitary transformation. This has the advantage that the equations are more readily deduced for any order of the perturbation, and the necessary integrations over the velocity distribution may then be finally made by inserting the spatial Fourier transform onto the cavity mode by analogy with the time-dependent iteration procedure. This effective-rate-constant approach would be of even greater utility in deducing the various coupled equations involved in Zeeman lasers, particularly in the case of an axial magnetic field, but we shall reserve such an application for a future account.

2. DIRECT ITERATION PROCEDURE

A. First-Order Theory

Referring to Fig. 1 and following the method developed by Lamb,⁷ we write the time-dependent equation for the density matrix as

$$\dot{\rho} = -i[\mathfrak{H}, \rho] - \frac{1}{2}[\Gamma\rho + \rho\Gamma], \quad (1)$$

with the Hamiltonian given by

$$\mathfrak{H} = \begin{pmatrix} \omega_a & V \\ V^* & \omega_b \end{pmatrix}, \quad (2)$$

and the time-dependent part of the perturbation as

$$\hbar V(t') = -\frac{1}{2}E(z-v(t-t'), t')\Delta^* \exp[-i(\nu_n t' + \phi)]. \quad (3)$$

Here Γ is a diagonal matrix representing the phenomenological decay of the atomic states, Δ is the matrix element of the transition $a \rightarrow b$, and ω_a and ω_b are the energies of the atomic states in angular frequency units. The rotating-wave approximation is automatically introduced by Eqs. (2) and (3). The equation for the macroscopic atomic polarization is then

$$P_n(t) = \Delta\rho_{ab}(t) + \Delta^*\rho_{ba}(t), \quad (4)$$

which must be transformed onto the spatial part of the cavity mode given by

$$U_n(z) = \sin K_n z, \quad (5)$$

and finally inserted into the conditional equation

$$\begin{aligned} & \frac{1}{2}\mathbf{1}[-i(2\nu_n\dot{E}_n + \nu\nu_n E_n/Q_n) + E_n 2\Omega_n(\Omega_n - (\nu_n + \phi))] \\ & \times \exp\{-i(\nu_n t + \phi)\} + \text{c.c.} \\ & = \frac{1}{2}\mathbf{1}(\nu^2/\epsilon_0)P_n(t) \exp\{-i(\nu_n t + \phi)\} + \text{c.c.}, \end{aligned} \quad (6)$$

where the unit vector $\mathbf{1}$ denotes the polarization of the emitted radiation.

Following the iteration procedure and changing the order of integration so as to integrate first over all values of t_0 , the time of excitation of a particular atomic state, we obtain the first-order result

$$\begin{aligned} \rho_{ab}^{(1)}(z, v, t) & = -i\Delta^*/(2\hbar)E(t) \exp[-i(\nu_n t + \phi)] \\ & \times \int_0^\infty \exp[-(\gamma - iT)\tau^I] N(z, t) U_n(z - v\tau^I) d\tau^I, \end{aligned} \quad (7)$$

where $N(z, t)$ is the excitation density, $\gamma = \frac{1}{2}(\gamma_a + \gamma_b)$, $T = (\nu_n - \omega_{ab})$ (cavity detuning), $\tau^I = t - t'$, and $\omega_{ab} = \omega_a - \omega_b$. Retaining only appropriate terms, the factor $U_n(z) U_n(z - v\tau^I)$, which occurs in the spatial Fourier projection onto the cavity mode, viz.,

$$P_n^{(1)}(v, t) = (2/L) \int_0^L P^{(1)}(z, v, t) U_n(z) dz, \quad (8)$$

may be reduced to the term

$$\frac{1}{2} \cos K v \tau^I. \quad (9)$$

This may now be substituted into Eq. (7) and the integrations over τ^I and z may be carried out.

Integrating finally over the assumed Maxwellian velocity distribution, we then obtain

$$\rho_{ab} = -i\Delta^*/(4\hbar)E(t) \exp[-i(\nu_n t + \phi)] N(z, t) I(v), \quad (10)$$

where

$$\begin{aligned} I(v) & = \frac{1}{\pi^{1/2}u} \int_{-\infty}^\infty \exp(-v^2/u^2) \\ & \times [(\gamma - iT - iKv)^{-1} + (\gamma - iT + iKv)^{-1}] dv. \end{aligned} \quad (11)$$

Here $K = 2\pi/\lambda$, and $\frac{1}{2}mu^2 = kT_a$, where T_a is the atomic temperature. The integrations are readily performed using the relationship^{8,9}

$$\frac{1}{\pi^{1/2}} \int_{-\infty}^\infty \frac{\exp(-t^2)}{\eta + i\xi + it} dt = -iZ(-\xi, \eta), \quad (12)$$

where η is positive, and $Z(\xi, i\eta)$ is the complex plasma

⁸ B. D. Fried and S. D. Conte, *The Plasma Dispersion Function—Hilbert Transform of the Gaussian* (Academic Press Inc., New York, 1961), p. 1.

⁹ H. R. Schlossberg and A. Javan, *Phys. Rev.* **150**, A267 (1966).

dispersion function.⁸ The contributions from the $\pm Kv$ terms are in fact equal, and we obtain the usual result

$$\rho_{ab}(t) = -(\Delta^*/\hbar Ku)^{\frac{1}{2}}[E(t)] \times \exp[-i(\nu_n t + \phi)] \bar{N}Z(\xi, \eta), \quad (13)$$

where $\xi = (\nu_n - \omega_{ab})/Ku$, $\eta = \gamma/Ku$, and \bar{N} is the excitation. Equation (12) is an important relationship between the velocity integral and the Z function, since

$$\begin{aligned} \rho_{ab}^{(3)}(z, v, t) = & i |\Delta|^2 \Delta^*/(8\hbar^3) E_n^3(t) \exp[-i(\nu_n t + \phi)] \\ & \times \iiint_0^\infty d\tau^I d\tau^{II} d\tau^{III} \exp[-(\gamma - iT)\tau^I] \exp(-\gamma_{a,b}\tau^{II}) \exp[-(\gamma \mp iT)\tau^{III}] \\ & \times N(z, t) U_n(z - v\tau^I) U_n(z - v(\tau^I + \tau^{II})) U_n(z - v(\tau^I + \tau^{II} + \tau^{III})), \end{aligned} \quad (14)$$

where $\tau^{II} = t' - t''$, $\tau^{III} = t' - t'''$, and the \mp sign in the exponent means that the integrations are performed with the negative sign in this term and then with the positive sign and the results added. The term $\exp(-\gamma_{a,b})$ means that γ_a is to be replaced by γ_b for an additional contribution from state b .

The product of the four sine functions now involved in Eq. (5) for the spatial Fourier projection onto the cavity mode may now be reduced to the expression

$$\begin{aligned} \frac{1}{8} [\cos Kv(\tau^I - \tau^{III}) + \cos Kv(\tau^I + \tau^{III}) \\ + \cos Kv(\tau^I + 2\tau^{II} + \tau^{III})], \end{aligned} \quad (15)$$

where as before we retain only significant terms in the expansion, and we consider only a single cavity mode. The exponential form of these expressions must now each be inserted into Eq. (14) and the integrations over z , τ^I , etc., performed, and then finally over the velocity distribution. Again we note that only the positive exponent in each cosine term need be considered, as the negative exponent gives the same contribution. The integrations over the velocity distribution are performed by expanding the various terms into partial fractions and using Eq. (12), together with the equation

$$\frac{1}{\pi^{1/2}} \int_{-\infty}^{\infty} \frac{\exp(-t^2)}{(\eta + i\xi + it)^2} dt = -iZ'(-\xi, \eta), \quad (16)$$

which is derived by differentiating both sides of Eq. (12) with respect to the real part of the complex argument. We shall merely indicate the results of these integrations, omitting the constant factors in Eqs. (14) and (15). In this respect we remember that a factor of 2 from the integral

$$\frac{2}{L} \int_0^L N(z, t) dz = 2\bar{N} \quad (17)$$

must finally be inserted.

the various integrals which occur in the higher-order perturbations can be resolved into partial fractions and integrated by this relationship or its derivatives.

B. Third-Order Results

In a similar way, carrying out the iteration to third order and simplifying, we obtain the basic third-order integrals

The term $\cos Kv(\tau^I - \tau^{III})$ gives the dominant third-order contributions

$$\begin{aligned} \{(\gamma_a Ku)^{-1}[-i(\gamma - iT)^{-1}Z(\xi, \eta) + \gamma^{-1}Z_i(\xi, \eta)] \\ + (\text{the same with } \gamma_b \text{ replacing } \gamma_a)\}. \end{aligned} \quad (18)$$

The $\cos Kv(\tau^I + \tau^{III})$ term gives the contributions

$$\begin{aligned} \{-\gamma_a^{-1}(Ku)^{-2}[Z'(\xi, \eta) + Z_r(\xi, \eta)/\xi] \\ + (\text{the same with } \gamma_b \text{ replacing } \gamma_a)\}. \end{aligned} \quad (19)$$

The $\cos Kv(\tau^I + 2\tau^{II} + \tau^{III})$ term is more complicated, but gives the following third-order contributions:

$$\begin{aligned} \{(\gamma - \gamma_a/2 - iT)^{-2}(2Ku)^{-1}[-iZ(0, \eta_a)] \\ + [\gamma_a - 2(\gamma - iT)]^{-1}(Ku)^{-2}[-Z'(\xi, \eta)] \\ + 2[\gamma_a - 2(\gamma - iT)^{-2}(Ku)^{-1}iZ(\xi, \eta)] \\ + (\text{same with } \gamma_b \text{ replacing } \gamma_a)\}, \end{aligned} \quad (20)$$

and

$$\begin{aligned} \{-\frac{1}{2}[\gamma_a - 2(\gamma - iT)]^{-1}(Ku)^{-2}Z(\xi, \eta)/\xi \\ - i[(\gamma - \gamma_a/2)^2 + T^2]^{-1}(2Ku)^{-1}Z(0, \eta_a) \\ - \frac{1}{2}[\gamma_a - 2(\gamma + iT)]^{-1}(Ku)^{-2}Z(-\xi, \eta)/(-\xi) \\ + (\text{same with } \gamma_b \text{ replacing } \gamma_a)\}. \end{aligned} \quad (21)$$

Here the parameter $\eta_a = \gamma_a/(2Ku)$, and similarly for γ_b . Z_r and Z_i represent the real and imaginary parts of the complex Z function.

The substitution of Eqs. (13) and (14) together with Eqs. (18) through (21) into Eq. (4) and finally into Eq. (6) gives the steady state of the laser oscillation to third order in the perturbation. It is apparent from Eqs. (18)–(21) that, in general, all terms must be considered in the expression for the saturation coefficient β of the laser oscillation, for which the real part of these expressions is required. The contributions from the spatial transform factors $\cos Kv(\tau^I + \tau^{III})$ and $\cos Kv(\tau^I + 2\tau^{II} + \tau^{III})$ are small when η is quite small,

but may become significant at values of η around 0.1 or more, and which may be encountered in practice. This applies particularly to the contribution given by Eq. (19). The results in Eqs. (20) and (21) due to the $\cos Kv(\tau^I + 2\tau^{II} + \tau^{III})$ factor are seen to depend on the individual decay constants γ_a and γ_b . Some cancellation between the various contributions is apparent, however, and it will appear later that the over-all contribution from this term will be small, though possibly not entirely negligible in all cases. The various expressions in Eqs. (20) and (21) also vary more rapidly with cavity detuning T , due to the mutual interference

between the Doppler-shifted traveling waves involved in them, and which is least around $T=0$. Nevertheless, the over-all curve of the contribution to the saturation coefficient β turns out to be a relatively flat function of cavity detuning T due to the cancellation between the various contributions. A similar behavior will be made evident in the similar fifth-order contributions which involve γ_a and γ_b explicitly.

C. Fifth-Order Terms

Proceeding to a fifth-order iteration of Eqs. (1) through (3) and simplifying, we obtain the equation

$$\begin{aligned} \rho_{ab}^{(5)}(z, v, t) = & -i |\Delta|^4 \Delta^* / (32\hbar^5) E_n^{(5)}(t) \exp[-i(\nu_n t + \phi)] \\ & \times \int_0^\infty d\tau^I d\tau^{II} d\tau^{III} d\tau^{IV} d\tau^V \exp[-(\gamma - iT)\tau^I] \exp(-\gamma_a \tau^{II}) \exp[-(\gamma \mp iT)\tau^{III}] \\ & \times \exp(-\gamma_a \tau^{IV}) \exp[-(\gamma \mp iT)\tau^V] N(z, t) U_n(z - v\tau^I) \\ & \times U_n(z - v(\tau^I + \tau^{II})) U_n(z - v(\tau^I + \tau^{II} + \tau^{III})) U_n(z - v(\tau^I + \tau^{II} + \tau^{III} + \tau^{IV})) \\ & \times U_n(z - v(\tau^I + \tau^{II} + \tau^{III} + \tau^{IV} + \tau^V)). \end{aligned} \quad (22)$$

All combinations of the plus and minus signs in Eq. (22) must be taken together with all the indicated replacements of γ_a by γ_b in the τ^{II} and τ^{IV} exponents, and which must be combined in all ways. The product of the six sine functions which now occurs in the spatial projection onto the cavity mode given by Eq. (8) may again be reduced to the following significant terms for our single cavity mode; these are

$$\frac{1}{3^{\frac{1}{2}}} [\cos Kv(\tau^I + \tau^{III} + \tau^V) + \cos Kv(\tau^V - \tau^I - \tau^{III}) + \cos Kv(\tau^V + \tau^I - \tau^{III}) + \cos Kv(\tau^{III} + \tau^V - \tau^I)], \quad (23)$$

which by analogy with the similar third-order contributions not involving τ^{II} may be expected to give the dominant fifth order contribution, and also the terms

$$\begin{aligned} \frac{1}{3^{\frac{1}{2}}} [\cos Kv(\tau^I + 2\tau^{II} + \tau^{III} + \tau^V) + \cos Kv(\tau^V - \tau^I - 2\tau^{II} - \tau^{III}) + \cos Kv(\tau^I + \tau^{III} + 2\tau^{IV} + \tau^V) \\ + \cos Kv(\tau^I - \tau^{III} - 2\tau^{IV} - \tau^V) + \cos Kv(\tau^I + 2\tau^{II} + \tau^{III} + 2\tau^{IV} + \tau^V) + \cos Kv(\tau^I + 2\tau^{II} + 3\tau^{III} + 2\tau^{IV} + \tau^V)], \end{aligned} \quad (24)$$

which by comparison with the term $\tau^I + 2\tau^{II} + \tau^{III}$ encountered in the third-order results, may be expected to give terms involving sharp resonances around line center, but with cancellation effects leading to a small over-all contribution from such fifth-order terms.

The various terms in Eqs. (23) and (24) must now be considered separately by substituting them into Eq. (22) and performing the simple integrations over z and over the τ factors as before. The results are then integrated over the velocity distribution by resolving the resultant terms into partial fractions and then using Eqs. (12) and (16), together with the additional relationship

$$\frac{1}{\pi^{1/2}} \int_{-\infty}^{\infty} \frac{\exp(-\xi^2)}{(\eta + i\xi + it)^3} dt = \frac{1}{2} i Z''(-\xi, \eta), \quad (25)$$

where the primes again denote the second derivative of the Z function with respect to the real part of the complex argument. There is no difficulty in principle apart from the number of terms which must now be considered.

Thus substituting Eq. (23) into Eq. (22) and performing the integrations we obtain the following contributions from the various velocity integrals which occur. These results are given for γ_a in both the τ^{II} and τ^{IV} exponents in Eq. (22), and those for the γ_b substitutions are readily deduced in this case since these terms are not involved in the velocity integrals deduced from Eq. (23). We indicate the integral involved in Eq. (22) by the signs of the iT terms involved in the τ^{III} and τ^V exponents, viz., $(-, -)$ etc., and we omit the constant term involved in all the integrals.

$\cos Kv(\tau^{III} + \tau^V - \tau^I)$ term:

$$(-, -) \begin{cases} -(i/2)(\gamma - iT)^{-2}(Ku)^{-1}Z(\xi, \eta) \\ -(\frac{1}{2})(\gamma - iT)^{-1}(Ku)^{-2}Z'(\xi, \eta), \end{cases} \quad (26)$$

$$(+, +) \begin{cases} -(i/4)\gamma^{-2}(Ku)^{-1}(Z(\xi, \eta) + Z(-\xi, \eta)) \\ -(2\gamma)^{-1}(Ku)^{-2}Z'(-\xi, \eta), \end{cases} \quad (27)$$

$$(-, +) \text{ and } (+, -) \begin{cases} -(i/4)\gamma^{-1}(\gamma - iT)^{-1}(Ku)^{-1}Z(\xi, \eta) \\ -(4\gamma)^{-1}(Ku)^{-2}[Z(-\xi, \eta)/(-\xi) + (\gamma - iT)^{-1}Z(\xi, \eta)/\xi]. \end{cases} \quad (28)$$

$\cos Kv(\tau^V + \tau^I - \tau^{III})$ term:

$$(-, -) \begin{cases} -(i/2)(\gamma - iT)^{-2}(Ku)^{-1}Z(\xi, \eta) \\ -(\frac{1}{2})(\gamma - iT)^{-1}(Ku)^{-2}Z'(\xi, \eta), \end{cases} \quad (29)$$

$$(+, +) \begin{cases} -(i/4)\gamma^{-1}(\gamma + iT)^{-1}(Ku)^{-1}Z(-\xi, \eta) \\ -(\frac{1}{4})(Ku)^{-2}[\gamma^{-1}Z(\xi, \eta)/\xi + (\gamma + iT)^{-1}Z(-\xi, \eta)/(-\xi)], \end{cases} \quad (30)$$

$$(-, +) \begin{cases} -(i/4)\gamma^{-1}(\gamma - iT)^{-1}(Ku)^{-1}Z(\xi, \eta) \\ -(\frac{1}{4})(Ku)^{-2}[\gamma^{-1}Z(-\xi, \eta)/\xi + (\gamma - iT)^{-1}Z(\xi, \eta)/\xi], \end{cases} \quad (31)$$

$$(+, -) \begin{cases} -(i/2)\gamma^{-2}(Ku)^{-1}Z(-\xi, \eta) \\ -(\frac{1}{2})\gamma^{-1}(Ku)^{-2}Z'(\xi, \eta). \end{cases} \quad (32)$$

$\cos Kv(\tau^V - \tau^I - \tau^{III})$ term:

$$(-, -) \begin{cases} -(i/2)(\gamma - iT)^{-2}(Ku)^{-1}Z(\xi, \eta) \\ -(\frac{1}{2})(\gamma - iT)^{-1}(Ku)^{-2}Z'(\xi, \eta), \end{cases} \quad (33)$$

$$(+, +) \begin{cases} -(i/4)\gamma^{-1}(\gamma + iT)^{-1}(Ku)^{-1}Z(-\xi, \eta) \\ -(\frac{1}{4})(Ku)^{-2}[\gamma^{-1}Z(\xi, \eta)/\xi + (\gamma + iT)^{-1}Z(-\xi, \eta)/(-\xi)], \end{cases} \quad (34)$$

$$(-, +) \begin{cases} -(i/4)\gamma^{-2}(Ku)^{-1}[Z(\xi, \eta) + Z(-\xi, \eta)] \\ -(\frac{1}{2})\gamma^{-1}(Ku)^{-2}Z'(\xi, \eta), \end{cases} \quad (35)$$

$$(+, -) \begin{cases} -(i/4)\gamma^{-1}(\gamma - iT)^{-1}(Ku)^{-1}Z(\xi, \eta) \\ -(\frac{1}{4})(Ku)^{-2}[\gamma^{-1}Z(-\xi, \eta)/(-\xi) + (\gamma - iT)^{-1}Z(\xi, \eta)/\xi]. \end{cases} \quad (36)$$

$\cos(\tau^I + \tau^{III} + \tau^V)$ term:

$$(-, -) \quad (i/2)(Ku)^{-3}Z''(\xi, \eta), \quad (37)$$

$$(+, +) \begin{cases} (i/4)T^{-1}(Ku)^{-2}[Z(\xi, \eta)/\xi + Z(-\xi, \eta)/(-\xi)] \\ -(i/2)T^{-1}(Ku)^{-2}Z'(-\xi, \eta), \end{cases} \quad (38)$$

$$(-, +) \text{ and } (+, -) \begin{cases} -(i/4)T^{-1}(Ku)^{-2}[Z(\xi, \eta)/\xi + Z(-\xi, \eta)/(-\xi)] \\ (i/2)T^{-1}(Ku)^{-2}Z'(\xi, \eta). \end{cases} \quad (39)$$

Since these integrations do not involve the exponents τ^{II} and τ^{IV} , the required interchanges of γ_a and γ_b in Eq. (22) simply multiply all the above results by the factor $(2\gamma/\gamma_a\gamma_b)^2$.

The same procedure has been carried out for all the terms in Eq. (24), but the resulting equations are too numerous to write out in full. We shall content ourselves with writing down the results obtained for each such

term when the signs of the iT exponents in Eq. (22) are taken as $(-, -)$. These results will be useful later on when we consider the relative magnitudes of the fifth-order contributions.

Thus, the $\cos Kv(\tau^V - \tau^I - 2\tau^{II} - \tau^{III})$ and $\cos Kv(\tau^I - \tau^{III} - 2\tau^{IV} - \tau^V)$ terms each give the contributions

$$\begin{aligned} & -(i/4)(\gamma - iT)^{-2}(\gamma_a + 2(\gamma - iT))^{-1}(Ku)^{-1}Z(\xi, \eta), \\ & -i(\gamma - \gamma_a/2 - iT)^{-2}(\gamma + \gamma_a/2 - iT)^{-1}(2Ku)^{-1}Z(0, \eta_a), \\ & -(i/2)(\gamma - iT)^{-1}(\gamma_a - 2(\gamma - iT))^{-1}(Ku)^{-2}Z'(\xi, \eta), \\ & \frac{-i[\gamma_a - 6(\gamma - iT)]}{4(\gamma - iT)^2(\gamma_a - 2(\gamma - iT))^2Ku}Z(\xi, \eta). \end{aligned} \quad (40)$$

The $\cos Kv(\tau^I + 2\tau^{II} + \tau^{III} + \tau^V)$ and $\cos Kv(\tau^I + \tau^{III} + 2\tau^{IV} + \tau^V)$ terms each give the contributions

$$\begin{aligned} & -i(\gamma - \gamma_a/2 - iT)^{-3}(2Ku)^{-1}Z(0, \eta_a), \\ & -4i(\gamma_a - 2(\gamma - iT))^{-3}(Ku)^{-1}Z(\xi, \eta), \\ & 2(\gamma_a - 2(\gamma - iT))^{-2}(Ku)^{-2}Z'(\xi, \eta), \\ & (i/2)(\gamma_a - 2(\gamma - iT))^{-1}(Ku)^{-3}Z''(\xi, \eta). \end{aligned} \quad (41)$$

The required interchanges of γ_a and γ_b in Eq. (22) are then obtained by replacing γ_a by γ_b throughout and multiplying the sum of the results by $2\gamma/\gamma_a\gamma_b$.

Similarly, the $\cos Kv(\tau^I + 2\tau^{II} + \tau^{III} + 2\tau^{IV} + \tau^V)$ term, with γ_a in both τ^{II} and τ^{IV} exponents, gives the results

$$\begin{aligned} & -8(2(\gamma - iT) - \gamma_a)^{-3}(2Ku)^{-2}Z'(0, \eta_a), \\ & 24i(2(\gamma - iT) - \gamma_a)^{-4}(2Ku)^{-1}Z(0, \eta_a), \\ & (i/2)(\gamma_a - 2(\gamma - iT))^{-2}(Ku)^{-3}Z''(\xi, \eta), \\ & 4(\gamma_a - 2(\gamma - iT))^{-3}(Ku)^{-2}Z'(\xi, \eta), \\ & -12i(\gamma - 2(\gamma - iT))^{-4}(Ku)^{-1}Z(\xi, \eta), \end{aligned} \quad (42)$$

while with γ_b in the τ^{II} exponent and γ_a in the τ^{IV} exponent, and vice versa, we obtain the results

$$\begin{aligned} & i(\gamma_b - \gamma_a)^{-1}(\gamma - \gamma_b/2 - iT)^{-3}(2Ku)^{-1}Z(0, \eta_b), \\ & i(\gamma_a - \gamma_b)^{-1}(\gamma - \gamma_a/2 - iT)^{-3}(2Ku)^{-1}Z(0, \eta_a), \\ & (i/2)(\gamma_b - 2(\gamma - iT))^{-1}(\gamma_a - 2(\gamma - iT))^{-1}(Ku)^{-3}Z''(\xi, \eta), \\ & -2(\gamma_a - 2(\gamma - iT))^{-1}(\gamma_b - 2(\gamma - iT))^{-1}[(\gamma_b - 2(\gamma - iT))^{-1} + (\gamma_a - 2(\gamma - iT))^{-1}](Ku)^{-2}Z'(\xi, \eta), \\ & -4i(\gamma_a - 2(\gamma - iT))^{-1}(\gamma_b - 2(\gamma - iT))^{-1}[(\gamma_b - 2(\gamma - iT))^{-2} \\ & + (\gamma_b - 2(\gamma - iT))^{-1}(\gamma_a - 2(\gamma - iT))^{-1} + (\gamma_a - 2(\gamma - iT))^{-2}](Ku)^{-1}Z(\xi, \eta). \end{aligned} \quad (43)$$

Finally the $\cos Kv(\tau^I + 2\tau^{II} + 3\tau^{III} + 2\tau^{IV} + \tau^V)$ term gives the similar contributions

$$\begin{aligned} & -27i(2(\gamma - iT))^{-2}(3\gamma_a - 2(\gamma - iT))^{-2}(3Ku)^{-1}Z(\xi/3, \eta/3), \\ & 2(\gamma_a - 2(\gamma - iT))^{-2}(\gamma - iT)^{-1}(Ku)^{-1}Z'(\xi, \eta), \\ & -8(2(\gamma - iT) - \gamma_a)^{-2}(2(\gamma - iT) - 3\gamma_a)^{-1}(2Ku)^{-2}Z'(0, \eta_a), \\ & (i/4)(3\gamma_a - 14\gamma)(\gamma_a - 2(\gamma - iT))^{-3}(\gamma - iT)^{-2}(Ku)^{-1}Z(\xi, \eta), \\ & 8i(10(\gamma - iT) - 9\gamma_a)(2(\gamma - iT) - \gamma_a)^{-3}(2(\gamma - iT) - 3\gamma_a)^{-2}(2Ku)^{-1}Z(0, \eta_a), \end{aligned} \quad (44)$$

and

$$\begin{aligned} & 8i(\gamma_a - \gamma_b)^{-1}(2(\gamma - iT) - \gamma_a)^{-2}(2(\gamma - iT) - 3\gamma_a)^{-1}(2Ku)^{-1}Z(0, \eta_a), \\ & 8i(\gamma_b - \gamma_a)^{-1}(2(\gamma - iT) - \gamma_b)^{-2}(2(\gamma - iT) - 3\gamma_b)^{-1}(2Ku)^{-1}Z(0, \eta_b), \\ & -27i(2(\gamma - iT))^{-2}(3\gamma_a - 2(\gamma - iT))^{-1}(3\gamma_b - 2(\gamma - iT))^{-1}(3Ku)^{-1}Z(-\xi/3, \eta/3), \\ & (2(\gamma - iT))^{-1}(\gamma_a - 2(\gamma - iT))^{-1}(\gamma_b - 2(\gamma - iT))^{-1}(Ku)^{-2}Z'(\xi, \eta), \\ & (i/2)(\gamma_a - 2(\gamma - iT))^{-1}(\gamma_b - 2(\gamma - iT))^{-1}(2(\gamma - iT))^{-1} \\ & [3(2(\gamma - iT))^{-1} - 2(\gamma_b - 2(\gamma - iT))^{-1} - 2(\gamma_a - 2(\gamma - iT))^{-1}](Ku)^{-1}Z(\xi, \eta). \end{aligned} \quad (45)$$

The interchanges of γ_a and γ_b now require the substitution of γ_b for γ_a in Eqs. (42) and (44), and the multiplication of Eqs. (43) and (45) by the factor 2.

D. Extension to Higher Orders

A comparison of the third- and fifth-order results for $\rho_{ab}(z, v, t)$, given by Eqs. (14) and (22), respectively, indicates that the basic seventh-order integral for this component of the density matrix will be given by

$$\begin{aligned} \rho_{ab}^{(7)}(z, v, t) = & i |\Delta|^6 \Delta^* / (128 \hbar^7) E_n^7 \exp[-i(v_n t + \phi)] \\ & \times \int_0^\infty d\tau^I d\tau^{II} d\tau^{III} d\tau^{IV} d\tau^V d\tau^{VI} d\tau^{VII} \exp[-(\gamma - iT)\tau^I] \exp(-\gamma_{a,b}\tau^{II}) \\ & \times \exp[-(\gamma \mp iT)\tau^{III}] \exp(-\gamma_{a,b}\tau^{IV}) \exp[-(\gamma \mp iT)\tau^V] \exp(-\gamma_{a,b}\tau^{VI}) \\ & \times \exp[-(\gamma \mp iT)\tau^{VII}] N(z, t) U_n(z - v\tau^I) \cdots \\ & \times U_n(z - v(\tau^I + \tau^{II} + \tau^{III} + \tau^{IV} + \tau^V + \tau^{VI} + \tau^{VII})), \end{aligned} \quad (46)$$

and similarly for the higher orders. The expansion of the eight sine functions now involved in the spatial Fourier projection onto the cavity mode represents a tedious problem. It can, however, be circumvented by applying the discussion given by Lamb⁷ on the interaction between the atom and the various running waves at the different times t' , t'' , etc.

Thus, if we consider the fifth-order problem again, the accumulated Doppler phase angle of the integrals in Eq. (22) will be given by

$$\begin{aligned} & \pm K v \tau^I \pm K v (\tau^I + \tau^{II}) \pm K v (\tau^I + \tau^{II} + \tau^{III}) \\ & \pm K v (\tau^I + \tau^{II} + \tau^{III} + \tau^{IV}) \\ & \pm K v (\tau^I + \tau^{II} + \tau^{III} + \tau^{IV} + \tau^V). \end{aligned} \quad (47)$$

Not all combinations of the signs, however, lead to a finite value of the spatial Fourier projection, and it follows that the effective combinations are those involving interactions with two running waves to the left (right) and three running waves to the right (left). This follows from the signs required of the propagation constants, in order that the integral over z may give a finite contribution. Thus if we take the sign combination in Eq. (47) as

$$\begin{aligned} & \pm K v [\tau^I + (\tau^I + \tau^{II}) + (\tau^I + \tau^{II} + \tau^{III}) \\ & - (\tau^I + \tau^{II} + \tau^{III} + \tau^{IV}) - (\tau^I + \tau^{II} + \tau^{III} + \tau^{IV} + \tau^V)], \end{aligned} \quad (48)$$

this then reduces to

$$\pm K v (\tau^I - \tau^{III} - 2\tau^{IV} - \tau^V), \quad (49)$$

and is one of the terms in Eq. (24). Similarly, all the other terms in Eqs. (23) and (24) may be obtained by this procedure, which represents a great simplification of the method by which these projections were originally obtained.

The extension of these considerations to the seventh and higher orders follows directly. Here an effective interaction would be with four right (left) running waves and three left (right) running waves. A typical

term in the spatial Fourier projection of the seventh-order atomic polarization onto the cavity mode would then be

$$(1/128) \cos K v (\tau^I + \tau^{III} - \tau^V + \tau^{VII}), \quad (50)$$

and similarly for all other such combinations. The integration of Eq. (46) may now be carried out as before, and the seventh order perturbation result presents no difficulty. For the present, however, we shall only consider the fifth-order contributions.

3. DISCUSSION OF THE RESULTS

The fifth-order terms must now be collated and substituted into Eq. (22) and then used in Eqs. (4) and (6), together with the first- and third-order contributions to the atomic polarization, to give the steady state of the laser oscillation. We shall confine our remarks to a discussion of the steady-state intensity to fifth order in the perturbation, the equation for which may then be written as

$$\dot{E} = \alpha E - \beta E^3 + \psi E^5. \quad (51)$$

The results of Sec. 2 may also be applied to determine the frequency of the laser oscillations, but we shall not discuss this any further.

Expressing the intensity in terms of the parameter

$$I = |\Delta|^2 E^2 / (\hbar^2 \gamma_a \gamma_b), \quad (52)$$

and omitting a constant factor of $v |\Delta|^2 \bar{N} / (2\epsilon_0 \hbar K u)$ in all the parameters, we then obtain the expressions

$$\alpha = Z_i(\xi, \eta) - \eta_i^{-1} Z_i(0, \eta), \quad (53)$$

$$\begin{aligned} \beta_1 = & \frac{1}{16} \text{Im} [2\gamma(\gamma - iT)^{-1} Z(\xi, \eta) + 2Z(\xi, \eta) \\ & - i2\eta(Z'(\xi, \eta) + Z(\xi, \eta)/\xi)], \end{aligned} \quad (54)$$

$$\begin{aligned} \beta_2 = & \frac{1}{16} \text{Im} \gamma_a \gamma_b \left[\frac{1}{2} (\gamma_b/2 - iT)^{-2} Z(0, \eta_a) \right. \\ & + i(Ku)^{-1} (\gamma_b - i2T)^{-1} (Z'(\xi, \eta) + Z(\xi, \eta)/\xi) \\ & - 2(\gamma_b - i2T)^{-2} Z(\xi, \eta) + \frac{1}{2} ((\gamma_b/2)^2 + T^2)^{-1} Z(0, \eta_a) \\ & \left. + (\text{same with an interchange of } \gamma_a \text{ and } \gamma_b) \right]. \end{aligned} \quad (55)$$

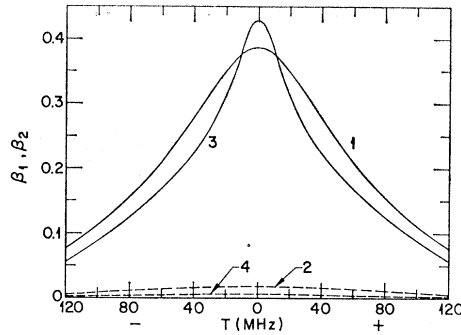


FIG. 2. Variation of β_1 (full curves) and β_2 (dashed curves) as a function of cavity detuning T . Curves 1 and 2, $\gamma=50$, $\gamma_b=75$, $\gamma_a=25$. Curves 3 and 4, $\gamma=20$, $\gamma_b=30$, $\gamma_a=10$. $Ku=100$ for all curves. All values expressed in MHz.

Here η_t represents the relative excitation or threshold parameter of the laser, and we have put $\beta=\beta_1+\beta_2$, where β_2 is the contribution from the $(\tau^I+2\tau^{II}+\tau^{III})$ term given by Eqs. (20) and (21). The relative magnitudes of β_1 and β_2 may be ascertained from their values at $T=0$ which is a maxima for both curves. Taking $\eta=0.2$, $Ku=100$ MHz, $\gamma_a=10$ MHz, and $\gamma_b=3\gamma_a$, we obtain the values $\beta_1=0.359+0.071=0.430$, where we have separated the two contributions from the

$$\cos(\tau^I-\tau^{III}) \quad \text{and} \quad \cos(\tau^I+\tau^{III})$$

terms in Eq. 15, and $\beta_2=0.0073$. Similarly, for $\eta=0.5$, $Ku=100$ MHz and $\gamma_b=3\gamma_a$, we obtain the values $\beta_1=0.273+0.113=0.386$, and $\beta_2=0.019$. Curves of β_1 and β_2 as functions of cavity detuning, deduced from Eqs. (54) and (55) are shown in Fig. 2, for the above values of the parameters. The β_2 curves are relatively flat functions of cavity detuning T , and give a small contribution to the saturation coefficient β for the values of η used here. It also follows from Eq. (55) that $\beta_2 \rightarrow 0$ as $\eta \rightarrow 0$.

Similarly, we may write $\psi=\psi_1+\psi_2$, where ψ_1 is the fifth-order contribution due to the terms in Eq. (23) and ψ_2 that due to the terms in Eq. (24). We then obtain the result

$$\begin{aligned} \psi_1 = & (1/128) \operatorname{Im}[3\gamma^2(\gamma-iT)^{-2}Z(\xi, \eta) \\ & -i3\eta\gamma(\gamma-iT)^{-1}(Z'(\xi, \eta)+Z(\xi, \eta)/\xi) \\ & -i3\eta Z'(\xi, \eta)+3\gamma(\gamma-iT)^{-1}Z(\xi, \eta) \\ & +3Z(\xi, \eta)-i3\eta Z(\xi, \eta)/\xi \\ & +i\eta^2 Z''(\xi, \eta)+i3\eta^2 Z'(\xi, \eta)/\xi]. \end{aligned} \quad (56)$$

The maximum value of this function occurs again at $T=0$, and its value may be deduced by applying the proper limiting procedure to Eq. (56), or it may be deduced directly from the values of the integrals in Eq. (22) which are all equal at $T=0$ for the same spatial

factor. In any event we find that

$$\psi_1 = \frac{1}{8^2} [3Z_i(0, \eta) - 3\eta Z'_r(0, \eta) + \eta^2 Z''(0, \eta)] \quad (57)$$

when $T=0$. Equation (57) then gives the value $\psi_1=0.164$ for $\eta=0.2$, and a limiting value of 0.166 is $\eta \rightarrow 0$. From Eqs. (54) and (57) we then find that the limiting value as $\eta \rightarrow 0$ for the ratio of ψ_1 to β is $\frac{2}{3}$. Figure 3 shows the values of ψ_1 as a function of cavity detuning T for the parameters $\eta=0.2$ and 0.5 with $Ku=100$ MHz.

As already indicated, the determination of ψ_2 as a function of T is quite complicated due to the numerous terms involved. This function, however, attains its maximum value at $T=0$, and this simplifies the deduction of its magnitude considerably, since the four integrals due to the sign permutations in Eq. (22) all have the same value for a given spatial factor $\cos Kv(\tau^I+2\tau^{II}+\tau^{III}+\tau^V)$, etc. The required ψ_2 contribution may then be deduced directly from Eqs. (22) and (24), or by multiplying the results already given in Eqs. (40) through (45) by the appropriate numerical factors as T goes to zero. Thus for the contributions from Eq. (40) we compute the result with the required interchange of γ_a and γ_b and multiply by the factor $(1/16)\gamma\gamma_a\gamma_b$, and similarly for all the other equations. Proceeding in this way we obtain the total contributions 0.03, 0.01, -0.08 , and 0.01, respectively, from Eqs. (40)–(45), which give a total ψ_2 value of -0.03 approximately at $T=0$. In these results the values $Ku=100$ MHz, $\eta=0.2$ and $\gamma_b=3\gamma_a$ have been used.

Comparing this value of ψ_2 with the value 0.164 of ψ_1 for these same parameters, we see that in general the effect of the ψ_2 terms will be small for the values of η and of the γ 's usually encountered in gas lasers, although some deviation from this may occur if the parameters get large. Even in the absence of any collision effects, the theory is thus reasonably consistent, and indicates that the curves of intensity versus cavity detuning will be only very slightly dependent on the β_2 and ψ_2 contributions. Also, if we introduce some collision broadening of the atomic response following the work of Szöke and Javan,¹⁰ these latter terms will become still smaller compared with β_1 and ψ_1 , due to the higher-order Lorentzian factors involved in Eq. (55) for β_2 and in Eqs. (40) through (45), which determine ψ_2 . In view of these results we shall neglect the β_2 and ψ_2 contributions in what follows.

From Eq. (51) the steady-state intensity of the laser is then given by

$$I = (\beta/2\psi) [1 - (1 - 4\alpha\psi/\beta^2)^{1/2}], \quad (58)$$

from which it follows that the condition

$$4\alpha\psi/\beta^2 < 1 \quad (59)$$

must be satisfied, otherwise the value deduced from

¹⁰ A. Szöke and A. Javan, Phys. Letters, 10, 521 (1963); Phys. Rev. 145, A137 (1966).

Eq. (58) is complex. For given values of ψ and β , this places a restriction on the value of α , and hence on the relative excitation parameter η_i , for which the fifth-order results are valid. It follows that the intensities deduced to fifth order will be more accurate if the inequality in Eq. (59) is satisfied by a fair margin, otherwise the perturbation treatment should really be taken to a higher order. When $\alpha\psi/\beta^2 \ll 1$, Eq. (58) may be approximated by the result

$$I = \alpha/\beta + \alpha^2\psi/\beta^3, \quad (60)$$

which shows that the intensity will be greater than that deduced from the usual third-order perturbation results. Figure 4 shows the intensity parameter $I = \alpha/\beta_1$ and that given by Eq. (58), with $\beta = \beta_1$ and $\psi = \psi_1$ as a function of cavity detuning T . The parameters $Ku = 100$ MHz and $\eta = 0.2$ have been used in the computations. The changes in the intensity curves, including the reduction in the Lamb dip, due to the fifth-order terms are evident. Relative excitations η_i of 1.2 and 1.05, respectively, were used in the deductions, and the inequality in Eq. (50) is then satisfied. It is apparent that the fifth-order terms are significant at values of η_i as low as 1.05 for which no Lamb dip occurs even in the third-order approximation with these parameters.

4. EFFECTIVE-RATE-CONSTANT APPROACH

The labor involved in deducing the higher-order perturbation equations by the time-dependent iteration method becomes even more tedious when more atomic levels and transitions must be considered, as in the theory of the Zeeman laser. It is thus advantageous, even for a two atomic level system, to remove the time dependence in the Hamiltonian by an appropriate time dependent diagonal unitary transformation.¹¹ The resulting equation of motion for the density matrix may then be solved directly or iterated, as before, the process then being much simpler since a number of terms are collated directly from the beginning. However, the method is no real substitute for the more rigorous one used in Sec. 2, and its success depends on the fact that

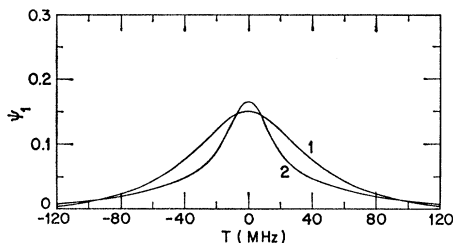


FIG. 3. Variation of ψ_1 versus cavity detuning T . Curve 1, $Ku = 100$, $\gamma = 50$. Curve 2, $Ku = 100$, $\gamma = 20$, expressed in MHz.

¹¹ L. R. Wilcox and W. E. Lamb, Jr., Phys. Rev. **119**, 1950 (1960); see also, W. E. Lamb, Jr., and T. M. Sanders, Jr., Phys. Rev. **119**, 1901 (1960).

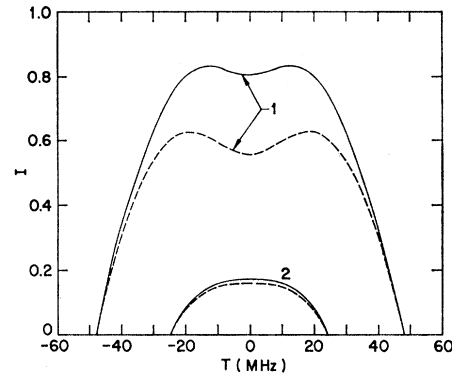


FIG. 4. Variation of laser intensity I , with cavity detuning T . $Ku = 100$ MHz, $\gamma = 20$ MHz. Curve 1, $\eta_i = 1.2$. Curve 2, $\eta_i = 1.05$. Full curves for the fifth-order approximation. Dashed curves for the usual third approximation.

the order of iterations may be monitored, and the velocity terms finally inserted using the known spatial projections of the atomic polarization onto the cavity mode. This is possible because the effective perturbation does not depend on t_0 , the time of excitation of any particular atomic state, and only the τ factors occur in the spatial projection. The method will be made clear by applying it to the two-level scheme shown in Fig. 1.

The Hamiltonian may now be written as

$$H = \begin{bmatrix} \omega_a & V \exp(-i\nu_n t) \\ V^* \exp(i\nu_n t) & \omega_b \end{bmatrix}, \quad (61)$$

and we require a diagonal unitary transformation U such that $U^\dagger H U$ is independent of the time.¹¹ The density matrix then transforms into

$$\sigma = U^\dagger \rho U, \quad (62)$$

and the equation of motion for σ is then given by

$$\dot{\sigma} = -(\Omega\sigma + \sigma\Omega^\dagger) + \tau, \quad (63)$$

where τ is a diagonal matrix representing the excitation rates of the various states, and where

$$\Omega = \frac{1}{2}\Gamma + iU^\dagger H U - \dot{U}^\dagger U \quad (64)$$

is now independent of time. The steady-state solution for σ is then given by the equation¹²

$$\Omega\sigma + \sigma\Omega^\dagger = \tau. \quad (65)$$

For the Hamiltonian given by Eq. (61) the required

¹² Here we formally identify the diagonal matrix τ with the excitation rate densities of Lamb's theory. These may be functions of the time, but will be relatively slowly varying compared with the time dependence of the optical perturbations, which has been removed by the unitary transformation. Equation (56) then follows from Eq. (54).

diagonal unitary transformation is given by

$$U(t) = \begin{bmatrix} 1 & 0 \\ 0 & \exp(i\nu_n t) \end{bmatrix}, \quad (66)$$

and from Eqs. (63)–(65) we obtain the equations

$$\begin{aligned} \gamma_a \sigma_{aa} - i(V^* \sigma_{ab} - V \sigma_{ba}) &= r_a, \\ \gamma_b \sigma_{bb} + i(V^* \sigma_{ab} - V \sigma_{ba}) &= r_b, \\ (a + b^*) \sigma_{ab} - iV(\sigma_{aa} - \sigma_{bb}) &= 0, \end{aligned} \quad (67)$$

where $a = \frac{1}{2}\gamma_a + i\omega_a$, $b = \frac{1}{2}\gamma_b + i(\omega_b + \nu_n)$. Equations (67) can be solved explicitly for σ_{ab} , but we may proceed by iteration which is better for more complicated level systems.

The first-order approximations are then given by

$$\sigma_{aa} - \sigma_{bb} = r_a/\gamma_a - r_b/\gamma_b \equiv N(z, t) \quad (68)$$

and

$$\sigma_{ab}^{(1)} = iVN(\gamma - iT)^{-1}. \quad (69)$$

The Doppler shift is now inserted using the spatial field factor $U_n(z)U_n(z - vt)$, the contributing term of which is $\frac{1}{2} \cos Kvt$ from Eq. (9). Noting that the denominator in Eq. (69) arises in the time-dependent iteration process from the integration of an exponential with a negative τ' exponent, and that the terms $\pm iKv$ give the same result, we may write

$$\sigma_{ab}^{(1)}(z, v, t) = (i/2)VN(z, t)[\gamma - i(T + Kv)]^{-1}. \quad (70)$$

Inserting the perturbation $V = -E(t)\Delta^*/(2\hbar)$, and integrating over z and over the velocity distribution as before, then gives the same result as Eq. (13) for the first-order contribution after transforming σ to ρ by Eq. (62).

Similarly, the third-order terms are deduced by substituting Eq. (69) into Eqs. (67), and we readily obtain the results

$$\sigma_{aa} - \sigma_{bb} = N[1 - |V|^2(\gamma_a^{-1} + \gamma_b^{-1})R], \quad (71)$$

$$\sigma_{ab}^{(3)} = -iVN/A[|V|^2(\gamma_a^{-1} + \gamma_b^{-1})R], \quad (72)$$

where $A = \gamma - iT$, and $R = A^{-1} + A^{*-1}$, an effective-rate constant. The insertion of the velocity variation is now done as before, using Eq. (15) for the various terms in the third-order spatial projection onto the cavity mode. Thus for the $\cos Kvt(\tau^I + \tau^{III})$, τ^I is associated with iVN/A , and τ^{III} with R in Eq. (72), which may then be written as

$$\begin{aligned} \sigma_{ab}^{(3)} &= -\frac{iVN}{\gamma - iT - iKv} \\ &\times \left[|V|^2 \frac{2\gamma}{\gamma_a \gamma_b} (\gamma - iT - iKv)^{-1} + (\gamma + iT - iKv)^{-1} \right]. \end{aligned} \quad (73)$$

Equation (73) then gives exactly the same result as Eq. (19) after integrating over z and over the velocity distribution.

In like manner the fifth-order contribution is given by

$$\sigma_{ab}^{(5)} = (iVN/A) |V|^4 [(\gamma_a^{-1} + \gamma_b^{-1})R(\gamma_a^{-1} + \gamma_b^{-1})R], \quad (74)$$

$(\tau^I) \qquad \qquad \qquad (\tau^{II}) \quad (\tau^{IV}) \quad (\tau^V)$

where we indicate the terms belonging to the respective τ parameters concerned. The velocity variation is now incorporated in a similar way by using Eqs. (23) and (24) for the spatial projection with exactly the same results as before. This method represents a concise and convenient way of collecting the various contributions which arise in the higher-order perturbations. In fact Eqs. (67) may be solved explicitly to give

$$\sigma_{ab} = (iVN/A)[1 + (\gamma_a^{-1} + \gamma_b^{-1})|V|^2 R]^{-1}, \quad (75)$$

and by a binomial expansion with the order of the factors maintained as in Eq. (74), the results for any order of the perturbation are readily deduced for inclusion of the velocity variations. This result is then in agreement with that already given in Eq. (46) for the seventh-order perturbation result. The method also proves to be very convenient in evaluating the various contributions to the atomic polarization terms in the Zeeman laser whenever the required unitary transformation can be found.

5. CONCLUSIONS

A more exact integration over the atomic velocity distribution, of the various terms occurring in the third- and fifth-order projections of the atomic polarization onto a single cavity mode, has been carried out for a two-level laser transition. The results, which are valid for any ratio of natural linewidth or cavity detuning to the Doppler width, show that there is a fifth-order contribution ψ_1 , the shape of which does not depend explicitly on γ_a or γ_b but only on $\gamma = \frac{1}{2}(\gamma_a + \gamma_b)$, and which should be considered even at low levels of laser intensity. Such contributions will not be materially reduced when collision effects are included, since they contain atomic response functions similar to those in the expression for the more usual third-order saturation coefficient. They will thus contribute to the over-all shape of the intensity-versus-cavity-detuning curves, leading to a general increase in the laser intensity and to a reduction in the Lamb-dip phenomena. The inequality $4\alpha\psi/\beta^2 < 1$, where ψ is the fifth-order coefficient in the equation for the steady-state intensity, must be satisfied to a reasonable degree, otherwise the perturbation should be taken to a higher

order. This places a restriction on the level of relative excitation for which the fifth-order results are reasonably valid, and shows that in general the higher-order terms should be considered in any deduction of decay constants made by inserting empirical values into the theory so as to obtain agreement with curves of experimental data.

In addition to the ψ_1 component of the fifth-order term discussed above, there are additional contributions β_2 and ψ_2 to third and fifth order which involve sharp resonances at line center. This is particularly the case for the numerous fifth-order contributions, which have this tendency due to Doppler-interference effects. Cancellation effects, however, occur among the various terms, and the resulting curves of β_2 and ψ_2 as functions of cavity tuning appear to be relatively flat, and small in magnitude. This is certainly so for β_2 , whilst ψ_2 has at least been shown to be relatively small in general compared with ψ_1 for the value $T=0$. These terms would give rise to an explicit dependence of the intensity-versus-cavity-detuning curves on both γ_a and γ_b as was encountered in earlier work.⁶ However, the more exact results given here show that such terms will in general have only a small effect on the tuning curves for the values of parameters usually encountered in gas lasers. This statement is substantiated by the computations given for $\eta=0.2$ and $\gamma_b=3\gamma_a$, though any widely deviating case must of course be considered on its merits using the expressions given here. Collision effects should reduce these terms still further, due to the higher-order atomic response functions involved in the pertinent expressions. The theory, however, appears

to be reasonably consistent even in the absence of any appeal to collision processes, in that no serious discrepancy between the deductions and present experimental results are apparent when the totality of the numerous fifth-order terms in ψ_2 is considered. The result that β_2 and ψ_2 may in general be considered small in relation to β_1 and ψ_1 , respectively, results in a considerable simplification of the fifth- and higher-order perturbation deductions, and such an approximation should be used whenever possible.

The effective-rate-constant approach represents a concise way of dealing with these higher-order perturbations, and leads to the same results as the more rigorous, but more tedious time-dependent iteration procedure, when used in the correct way. It may be applied with even greater utility to the more complicated atomic-level schemes and transitions encountered in Zeeman lasers, and may be used to discuss the effects of axial magnetic fields on all laser transitions so far investigated. The situation with a transverse magnetic field is, however, more complicated, and the required unitary transformation can only be found in the simpler cases, such as for a $J=1 \rightarrow 0$ transition. The higher-order perturbations are most likely of some significance in Zeeman lasers, particularly as regards mode competition and the stability of the oscillations, but we must defer any such application for the present.

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