ZnS:Si. This is contradictory to the experimental results. The agreement between the calculated and the observed Δg's seems fairly good. Although this is perhaps fortuitous because of the crudeness of the parameters used, the sign and the order of magnitude of the g shifts seems to be interpreted qualitatively on the theory based on our proposed model.

The observed spin densities, $\rho_s(0)$, are extremely large. We will tentatively compare the observed values with those of the s orbitals of the free atoms. Available values of $|\psi_s(0)|^2_{\text{free}}$ of neutral atoms[17,18] calculated from Hartree-Fock wave functions are also listed in Table II. The observed densities are approximately 60% of those of free atoms. Any quantitative discussion of these values could hardly be fruitful from (10) in our crude one-electron linear-combination-of-atomic-orbitals approximation. Quantitative calculations of the unpaired spin densities would require the detailed knowledge of wave function for unpaired electron and of the inner core polarization.

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Effects of Atomic Degeneracy and Cavity Anisotropy on the Behavior of a Gas Laser

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Expressions are developed in this paper which describe the behavior of a gas laser having generalized polarization characteristics. It is found that degeneracies of the atomic energy levels play an important part in determining the behavior of such a laser since significant terms occur in the nonlinear polarization which are attributable to an oscillatory mixing of these levels. As a result, it is found, for example, that for single-mode operation the field intensity is greatest for either plane or circular polarization depending upon whether a $\Delta j=\pm 1$ or $\Delta j=0$ atomic transition is involved in the laser action. For two-mode operation, on the other hand, the behavior depends in a complicated way both on the polarization states of the oscillations and on the degree of degeneracy of the energy levels. This behavior is discussed in a number of special cases.

I. INTRODUCTION

In this paper, we extend Lamb's theory of an optical maser[1] to cover systems involving degenerate atomic energy levels and optical oscillations having arbitrary states of polarization. Our treatment is specifically aimed at determining the output characteristics of gas lasers which utilize generalized anisotropic resonant cavities. Such lasers are of interest because of the unambiguous control of their frequency and polarization characteristics which is made possible by the use of nonresonant intra-cavity anisotropic components. In addition to forming the basis for a number of practical devices,[2,3] this control is quite useful in the study of the fundamental atomic phenomena governing laser behavior.

A recent theoretical and experimental study of the properties of a Fabry-Perot resonator containing an array of retardation plates and Faraday rotators has established that each longitudinal mode of such a cavity is split into two distinct resonances.[3] The frequency separation between these resonances is dependent upon the strength of the anisotropic effects and their preferred states of polarization are in general elliptical and orthogonal. The introduction of anisotropic losses into such a resonator can furthermore break down the orthogonality of the preferred polarization states and, in extreme cases, can restrict oscillation to a single polarization. In the discussion below, we will thus give particular attention to lasers whose oscillations belong to two distinct, but not necessarily orthogonal, states of polarization.

Our theoretical approach is quite similar to that of Lamb, the chief distinctions being our explicit treatment of the vectorial nature of the electromagnetic field and of the degeneracies of the atomic system. In the sections below, the reader will thus be referred to Lamb's work for more detailed discussions of some of the conditions of the problem and for treatment of those calculative details which are common to the two works. It should

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finally be noted that our assumption of complete degeneracy of the magnetic sublevels belonging to a given J^2 energy level implies the absence of magnetically induced anisotropies in the laser medium. This situation can usually be approached quite adequately in experiment by the use of a moderate amount of shielding to reduce the strength of the earth's magnetic field.

II. FIELD EQUATIONS

Using a model similar to that used by Lamb, we assume the electric field within the active medium to obey the wave equation

$$\text{curl curl}\mathbf{E}+\mu_0\frac{\partial}{\partial t}\boldsymbol{\sigma}\cdot\mathbf{E}+\mu_0\epsilon_0\frac{\partial^2}{\partial t^2}\mathbf{E}=-\mu_0\frac{\partial^2}{\partial t^2}\mathbf{P}, \quad (1)$$

where the anisotropic nature of the resonant cavity has been taken into account by the introduction of the fictional conductivity tensor $\boldsymbol{\sigma}$. As will be seen later, the nonlinear contribution to the induced polarization $\mathbf{P}$ is not necessarily parallel to $\mathbf{E}$. Thus, in general, neither of these vectors is parallel to $\boldsymbol{\sigma}\cdot\mathbf{E}$, and it would normally be necessary to consider the specific form of $\boldsymbol{\sigma}$. In order to avoid this considerable complication, we will assume that both the field and the induced polarization may be decomposed into two sets of oscillations, each of which is in one of the two polarization eigenstates $\mathbf{e}_a$ or $\mathbf{e}_b$ of the passive cavity. It is then possible to replace the conductivity tensor by its eigenvalues σ_a and σ_b. The validity of this approximation is dependent on the relative magnitudes of the cavity anisotropies and the nonlinearities of the medium. Its consequences will be discussed later. We thus assume that

$$\mathbf{E}=\sum_n[\mathbf{e}_aA_n{}^a(t)\sin K_n{}^az+\mathbf{e}_bA_n{}^b(t)\sin K_n{}^bz] \quad (2)$$

with

$$A_n{}^{a/b}(t)=E_n{}^{a/b}(t)\cos(\nu_n{}^{a/b}t+\phi_n{}^{a/b}) \quad (3)$$

and

$$K_n{}^{a/b}=n\pi/L^{a/b}, \quad (4)$$

where a/b means either a or b, and the effective cavity lengths $L^{a/b}$ for the two polarization states are, in general, different. In our approximation, we must project the various frequency components of the calculated induced polarization $\mathbf{P}$ along the chosen field directions. We thus use in Eq. (1)

$$\begin{aligned}\mathbf{P}^{\|}=\sum_n[&\mathbf{e}_a\mathbf{e}_a\cdot\mathbf{C}_n{}^a(t,z)\cos(\nu_n{}^at+\phi_n{}^a(t))+\mathbf{e}_b\mathbf{e}_b\cdot\mathbf{C}_n{}^b(t,z)\\&\times\cos(\nu_n{}^bt+\phi_n{}^b(t))+\mathbf{e}_a\mathbf{e}_a\cdot\mathbf{S}_n{}^a(t,z)\sin(\nu_n{}^at+\phi_n{}^a(t))\\&+\mathbf{e}_b\mathbf{e}_b\cdot S_n{}^b(t,z)\sin(\nu_n{}^bt+\phi_n{}^b(t))], \quad (5)\end{aligned}$$

where $\mathbf{C}_n{}^{a/b}$ and $\mathbf{S}_n{}^{a/b}$ are the in-phase and quadrature amplitudes of the polarization induced by $\mathbf{E}$.

Equations relating the component field amplitude $E_n{}^{a/b}(t)$ to $\mathbf{e}_{a/b}\cdot\mathbf{C}_n{}^{a/b}$ and $\mathbf{e}_{a/b}\cdot\mathbf{S}_n{}^{a/b}$ may be obtained by substituting Eqs. (2) and (5) into the wave equation and equating coefficients of like time-dependent trigonometric functions. In addition, the spatial dependence may be removed by multiplying each equation by $\sin K_n{}^{a/b}z$ and integrating over the appropriate cavity length.

We obtain

$$(\nu_n{}^{a/b}+\dot{\phi}_n{}^{a/b}-\Omega_n{}^{a/b})E_n{}^{a/b}=-(\nu/2\epsilon_0)\mathbf{e}_{a/b}\cdot\mathbf{C}_n{}^{a/b}(t), \quad (6)$$

and

$$\dot{E}_n{}^{a/b}+(\nu/2Q^{a/b})E_n{}^{a/b}=-(\nu/2\epsilon_0)\mathbf{e}_{a/b}\cdot\mathbf{S}_n{}^{a/b}(t), \quad (7)$$

where

$$\mathbf{C}_n{}^{a/b}(t)=\int_0^{L^{a/b}}\mathbf{C}_n{}^{a/b}(t,z)\sin K_n{}^{a/b}z\,dz, \quad (8)$$

$$\mathbf{S}_n{}^{a/b}(t)=\int_0^{L^{a/b}}\mathbf{S}_n{}^{a/b}(t,z)\sin K_n{}^{a/b}z\,dz, \quad (9)$$

$$\Omega_n{}^{a/b}=cK_n{}^{a/b}, \quad (10)$$

and

$$Q^{a/b}=\epsilon_0\nu/\sigma^{a/b}. \quad (11)$$

III. ATOMIC POLARIZATION

The quantities appropriate for substitution into the self-consistent-field equations are obtained by calculating, to third order, the polarization induced in the laser medium by a multifrequency optical field $\mathbf{E}(z,t)$. In this calculation it is assumed that two excited atomic levels, characterized by angular momentum quantum numbers j and j', interact with the optical field and that the J_z eigenstates associated with a given J^2 eigenstate are completely degenerate. Interatomic collision effects as well as population of the lower states (j') through spontaneous decay of the upper state (j) are neglected.

It is convenient, following Lamb's work,[1] to adopt a density matrix formulation in which the equation of motion

$$\dot{\rho}=-i[H,\rho]-\tfrac{1}{2}(\Gamma\rho+\rho\Gamma) \quad (12)$$

is solved separately for ensembles of atoms characterizing various portions of the atomic distribution, and the results are integrated over this distribution to yield the total polarization. In this equation ρ is the density matrix, H is the perturbed atomic Hamiltonian and, in the representation of unperturbed J^2, J_z eigenstates,

$$\Gamma_{mm';jj'}=\gamma_j\delta_{mm';jj'}, \quad (13)$$

where γ_j is the spontaneous decay constant of state j.

From the general definition of the density matrix it follows that, if the x axis is taken to be the axis of quantization, each atomic ensemble makes the contributions

$$P_x(z,t)=\sum_m \mathcal{P}_{mm;j'j}\rho_{mm;jj'}(z,t)+\text{conj}, \tag{14}$$

and

$$P_y(z,t)=\sum_m[\mathcal{P}_{m,m+1;j'j}\rho_{m+1,m;jj'}(z,t)+\mathcal{P}_{mm-1;j'j}\rho_{m-1,m;jj'}(z,t)]+\text{conj}. \tag{15}$$

to the Cartesian components of the atomic polarization. The quantity $\mathcal{P}_{mn;jj'}$ above designates the matrix element of the component of the atomic dipole moment operator either along or perpendicular to the quantitazation axis depending on whether $n=m$ or $n=m\pm1$, respectively.

Writing out Eq. (12) in component form yields

$$\dot{\rho}_{mn;jj}=-\gamma_j\rho_{mn;jj}+i\sum_p(V_{mp}\rho_{pn;j'j}-V'_{pn}\rho_{mp;jj'}), \tag{16}$$

and

$$\dot{\rho}_{mn;jj'}=(-i\omega-\gamma)\rho_{mn;jj'}+i\sum_p(V_{mp}\rho_{pn;j'j'}-V_{pn}\rho_{mp;jj}), \tag{17}$$

where

$$j'=j,\ j\pm1$$

$$\omega=W_j-W_{j'}>0, \tag{18}$$

$$\gamma=\tfrac{1}{2}(\gamma_j+\gamma_{j'}), \tag{19}$$

$$\mathcal{P}_{mn}=\mathcal{P}_{mn;jj'}, \tag{20}$$

$$\mathcal{P}_{mn}'=\mathcal{P}_{mn;j'j}, \tag{21}$$

$$\begin{aligned}\hbar V_{mn}(t)&=E_x^R(t)\mathcal{P}_{mn;jj'}\quad\text{if}\quad m=n\\&=E_y^R(t)\mathcal{P}_{mn;jj'}\quad\text{if}\quad m\neq n,\end{aligned} \tag{22}$$

and

$$\begin{aligned}\hbar V_{mn}'(t)&=E_x^R(t)\mathcal{P}_{mn;j'j}\quad\text{if}\quad m=n\\&=E_y^R(t)\mathcal{P}_{mn;j'j}\quad\text{if}\quad m\neq n.\end{aligned} \tag{23}$$

Here $\mathbf{E}^R(t)=E_x^R(t)\mathbf{i}+E_y^R(t)\mathbf{j}$ is the optical-frequency electric field at time t in the rest frame of atoms in the ensemble under consideration.

Assuming equal excitation of all m substates within a given j state, Eqs. (16) and (17) can now be solved by iteration for ensembles of atoms of velocity v excited to either angular momentum state at time t_0 and position z_0. For initial excitation to the upper state the solutions to the first three orders in the V's are

$$\rho_{mn;jj'}^{(1)}(j,z_0,t_0,v,t)=-\frac{i}{2j+1}\int_{t_0}^{t}dt'\,V_{mn}(t')\exp[(\gamma+i\omega)(t'-t)+\gamma_j(t_0-t')], \tag{24}$$

$$\begin{aligned}\rho_{mn;jj}^{(2)}(j,z_0,t_0,v,t)=-\frac{i}{2j+1}\int_{t_0}^{t}dt''\int_{t_0}^{t''}dt'''\,\exp[\gamma_j(t''-t)]\sum_p\{V_{mp}(t'')V_{pn}'(t''')\\\times\exp[(\gamma-i\omega)(t'''-t'')+\gamma_j(t_0-t''')]+V_{pn}'(t'')V_{mp}(t''')\exp[(\gamma+i\omega)(t'''-t'')+\gamma_j(t_0-t''')]\},\end{aligned} \tag{25}$$

and

$$\begin{aligned}\rho_{mn;jj'}^{(3)}(j,z_0,t_0,v,t)=\frac{i}{2j+1}\int_{t_0}^{t}dt'\int_{t_0}^{t'}dt''\int_{t_0}^{t''}dt'''\,\exp[(\gamma+i\omega)(t'-t)+\gamma_j(t_0-t''')]\{\exp[\gamma_{j'}(t''-t')]\\\times\sum_{pq}[V_{mp}(t')V_{pq}'(t'')V_{qn}(t''')\exp[(\gamma+i\omega)(t'''-t'')]+V_{mp}(t')V_{qn}(t'')V_{pq}'(t''')\\\times\exp[(\gamma-i\omega)(t'''-t'')]]+\exp[\gamma_j(t''-t')]\sum_{pq}[V_{pn}(t')V_{mq}(t'')V_{qp}'(t''')\exp[(\gamma-i\omega)(t'''-t'')]\\+V_{pn}(t')V_{qp}'(t'')V_{mq}(t''')\exp[(\gamma+i\omega)(t'''-t'')]]\}.\end{aligned} \tag{26}$$

The total polarization $\mathbf{P}(z,t)$ is obtained by first multiplying Eqs. (24) and (26) and the corresponding equations for initial lower state excitation by the appropriate excitation rate parameters and then adding the results. The composite equation is next integrated over all t_0 such that $(z-z_0)=v(t-t_0)$, and over a Maxwellian velocity distribution. The expression resulting from these integrations is finally substituted into Eqs. (14) and (15) to yield the Cartesian components of $\mathbf{P}(z,t)$. For an assumed optical-frequency electric field of the form

$$\mathbf{E}(z,t)=E_x(z,t)\mathbf{i}+E_y(z,t)\mathbf{j}, \tag{27}$$

where

$$E_x(z,t)=\sum_{\mu l}E_{\mu x}^l\cos(\nu_\mu^l t+\phi_{\mu x}^l)\sin K_\mu^l z \tag{28}$$

and

$$E_y(z,t)=\sum_{\mu l}E_{\mu y}^l\cos(\nu_\mu^l t+\phi_{\mu y}^l)\sin K_\mu^l z \tag{29}$$

with

$$K_\mu^l(z)=\mu\pi/L_l, \tag{30}$$

a calculation similar to that carried out by Lamb for finite atomic velocities leads, in the Doppler limit, to the following expressions for the spatial Fourier projections of the first- and third-order atomic polarizations.

$$P_{nx}{}^{(1)v}=-(2\hbar Ku)^{-1}[\sum_m|\mathcal{P}_{mm}|^2][\sum_{\mu l}E_{\mu x}{}^l\exp[-i(\nu_\mu{}^lt+\phi_{\mu x}{}^l)]N_{(n-\mu)}{}^{(v-l)}Z(\nu_\mu{}^l-\omega)]+\text{conj.} \quad (31)$$

and

$$P_{nx}{}^{(3)v}=\frac{i\pi^{1/2}}{32\hbar^3Ku}\sum_{\substack{\mu\rho\sigma\\ lst}}\{\mathfrak{D}(\nu_\rho{}^s-\bar\nu_{\mu\sigma}{}^{lt})[\mathfrak{D}_{j'}(\nu_\rho{}^s-\nu_\sigma{}^t)+\mathfrak{D}_j(\nu_\rho{}^s-\nu_\mu{}^l)]N_{(\mu-\rho+\sigma-n)}{}^{(l-s+t-v)}+\mathfrak{D}_{j'}(\nu_\rho{}^s-\nu_\sigma{}^t)$$
$$\times\mathfrak{D}(\omega-\tfrac12\nu_\mu{}^l+\tfrac12\nu_\rho{}^s-\nu_\sigma{}^t)N_{(\rho-\sigma+\mu-n)}{}^{(s-t+l-v)}+\mathfrak{D}_j(\nu_\rho{}^s-\nu_\mu{}^l)\mathfrak{D}(\omega-\tfrac12\nu_\sigma{}^t+\tfrac12\nu_\rho{}^s-\nu_\mu{}^l)N_{(\rho-\mu+\sigma-n)}{}^{(s-l+t-v)}\}$$
$$\times\exp[-i(\nu_\mu{}^l-\nu_\rho{}^s+\nu_\sigma{}^t)t]\{AE_{\mu x}{}^lE_{\rho x}{}^sE_{\sigma x}{}^t\exp[-i(\phi_{\mu x}{}^l-\phi_{\rho x}{}^s+\phi_{\sigma x}{}^t)]+BE_{\mu x}{}^lE_{\rho y}{}^sE_{\sigma y}{}^t$$
$$\times\exp[-i(\phi_{\mu x}{}^l-\phi_{\rho y}{}^s+\phi_{\sigma y}{}^t)]+CE_{\mu y}{}^lE_{\rho y}{}^sE_{\sigma x}{}^t\exp[-i(\phi_{\mu x}{}^l-\phi_{\rho y}{}^s+\phi_{\sigma x}{}^t)]$$
$$+DE_{\mu y}{}^lE_{\rho x}{}^sE_{\sigma y}{}^t\exp[-i(\phi_{\mu y}{}^l-\phi_{\rho x}{}^s+\phi_{\sigma y}{}^t)]\}+\text{conj.}, \quad (32)$$

where

$$P_{nx}{}^v=\frac{2}{L_v}\int_0^{L_v}dz\,P_x(z,t)\sin K_n{}^vz, \quad (33)$$

$$N_{(\mu-\rho+\sigma-n)}{}^{(l-s+t-v)}=\frac{2}{L_v}\int_0^{L_v}dz\,N(z,t)\times\cos[(K_\mu{}^l-K_\rho{}^s+K_\sigma{}^t-K_n{}^v)z], \quad (34)$$

$$N(z,t)=\Lambda_j(z,t)/(2j+1)\gamma_j-\Lambda_{j'}(z,t)/(2j'+1)\gamma_{j'}, \quad (35)$$

$$z(\nu-\omega)=iKu\int_0^\infty d\tau\exp[i(\nu-\omega)\tau-\gamma\tau-\tfrac14K^2u^2\tau^2], \quad (36)$$

$$\mathfrak{D}(\nu)=(\gamma+i\nu)^{-1}, \quad (37)$$

$$\mathfrak{D}_j(\nu)=(\gamma_j+i\nu)^{-1}, \quad (38)$$

$$\bar\nu_{\mu\sigma}{}^{lt}=\tfrac12(\nu_\mu{}^l+\nu_\sigma{}^t), \quad (39)$$

$$A=\sum_m|\mathcal{P}_{mm}|^4, \quad (40)$$

$$B=\sum_m|\mathcal{P}_{mm}|^2[|\mathcal{P}_{m+1,m}|^2+|\mathcal{P}_{m-1,m}|^2], \quad (41)$$

$$C=\sum_m|\mathcal{P}_{mm}|^2[|\mathcal{P}_{m,m-1}|^2+|\mathcal{P}_{m,m+1}|^2], \quad (42)$$

and

$$D=\sum_m[\mathcal{P}_{m,m}\mathcal{P}_{m,m-1}\mathcal{P}_{m-1,m-1}\mathcal{P}_{m-1,m}+\mathcal{P}_{m,m}\mathcal{P}_{m,m+1}\mathcal{P}_{m+1,m+1}\mathcal{P}_{m+1,m}]. \quad (43)$$

In the above expression $N(z,t)$ is the excitation density per m substate, and $\Lambda_j(z,t)$ and $\Lambda_{j'}(z,t)$ are the total numbers of excitations to the states j and j', respectively, per unit time per unit volume.

Expressions for $P_{ny}{}^{v(1)}$ and $P_{ny}{}^{v(3)}$ are obtained by interchaing x and y in Eqs. (31) and (32), respectively. The rather complex form of the optical-frequency electric field [Eq. (27)] was chosen so as to allow for the existence of simultaneously oscillating modes corresponding to different laser cavity lengths. As indicated above, this degree of generality is necessary for the treatment of systems using generalized anisotropic resonant cavities.

IV. AMPLITUDES AND FREQUENCIES IN AN ANISOTROPIC LASER

In order to obtain information about mode competition and pulling effects in an anisotropic laser, we now assume the existence of two modes having the same principle order number but corresponding to slightly different effective cavity lengths. Under these conditions the indices μ, ρ, σ, and n of Eqs. (31) and (32) are equal, while the indices l, s, t, and v can assume only the values 1 and 2. Letting $\mathbf{P}_n{}^1=\mathbf{P}_1$, $\mathbf{E}_n{}^1=\mathbf{E}_1$, $\nu_n{}^1=\nu_1$, $\nu_n{}^2=\nu_2$, and $N_{(n-n+n-n)}{}^{(l-s+t-v)}=N_{(l-s+t-v)}$ and dropping all terms with frequencies not equal to ν_1, Eq. (32) reduces to

$$P_{1x}{}^{(3)}=(i\pi^{1/2}/32\hbar^3Ku)\{Q_1[AE_{1x}{}^3+(B+C+D\exp2i\psi_1)\times E_{1x}E_{1y}{}^2]+A(M_{12}+M_{12}{}')E_{1x}E_{2x}{}^2+(M_{12}B+M_{12}{}'C)E_{1x}E_{2y}{}^2+[(M_{12}C+M_{12}{}'B)\times\exp i(\psi_1-\psi_2)+L(M_{12}+M_{12}{}')\exp i(\psi_1+\psi_2)]\times E_{2x}E_{2y}E_{1y}\}\exp-i(\nu_1t+\phi_{1x})+\text{conj.}, \quad (44)$$

where

$$Q_1=[\mathfrak{D}(0)+\mathfrak{D}(\omega-\nu_1)][\mathfrak{D}_j(0)+\mathfrak{D}_{j'}(0)]N_{10}, \quad (45)$$

$$M_{12}=\mathfrak{D}_{j'}(0)[\mathfrak{D}(\Delta)+\mathfrak{D}(\omega-\bar\nu)]N_{10}+\mathfrak{D}_j(2\Delta)[N_{10}\mathfrak{D}(\Delta)+N_{12}\mathfrak{D}(\omega-\nu_1)], \quad (46)$$

$$M_{12}{}'=\mathfrak{D}_j(0)[\mathfrak{D}(\Delta)+\mathfrak{D}(\omega-\bar\nu)]N_{10}+\mathfrak{D}_{j'}(2\Delta)[N_{10}\mathfrak{D}(\Delta)+N_{12}\mathfrak{D}(\omega-\nu_1)], \quad (47)$$

$$\Delta=\tfrac12(\nu_2-\nu_1), \quad (48)$$

$$\bar\nu=\bar\nu_{12}, \quad (49)$$

$$\psi_1=\phi_{1x}-\phi_{1y}, \quad (50)$$

$$\psi_2=\phi_{2x}-\phi_{2y}, \quad (51)$$

$$N_{10}=\frac{2}{L_1}\int_0^{L_1}dz\,N(z,t), \quad (52)$$

and

$$N_{12}=\frac{2}{L_1}\int_0^{L_1} dz\, N(z,t)\cos[2(K_2-K_1)z]. \tag{53}$$

A corresponding expression for $P_{1y}^{(3)}$ can be obtained by interchanging y and x in Eqs. (44) and (45). $P_{2x}^{(3)}$ is obtained by interchanging the subscripts 1 and 2.

It is clear from the definition of $\mathbf{P}_n$ given in Eq. (33) and the definitions of $\mathbf{C}_n^{a/b}(t,z)$ and $\mathbf{S}_n^{a/b}(t,z)$ given in Sec. II that if we identify ν_1 with ν_n^a and ν_2 with ν_n^b the in-phase and quadrature portions of $\mathbf{P}_1$ can be identified with $\mathbf{C}_n^a(t)$ and $\mathbf{S}_n^a(t)$ of Eqs. (8) and (9), and the corresponding portions of $\mathbf{P}_2$ can be identified with $\mathbf{C}_n^b(t)$ and $\mathbf{S}_n^b(t)$. Extracting the quadrature part of Eq. (44), we have

$$\begin{aligned}S_{1x}^{(3)}=(\pi^{1/2}/16\hbar^3Ku)\{&[AQ_1^r]E_{1x}^3+[(B+C)Q_1^r+D(Q_1^r\cos2\psi_1+Q_1^i\sin2\psi_1)]E_{1x}E_{1y}^2+[A(M_{12}^r+M_{12}'^r)]E_{1x}E_{2x}^2\\&+[BM_{12}^r+CM_{12}'^r]E_{1x}E_{2y}^2+[(CM_{12}^r+BM_{12}'^r)\cos(\psi_1-\psi_2)-(CM_{12}^i+BM_{12}'^i)\sin(\psi_1-\psi_2)\\&+D(M_{12}^r+M_{12}'^r)\cos(\psi_1+\psi_2)-D(M_{12}^i+M_{12}'^i)\sin(\psi_1+\psi_2)]E_{2x}E_{1y}E_{2y}\}, \end{aligned}\tag{54}$$

where

$$Q_1^r=\mathrm{Re}Q_1=(2/\gamma_j\gamma_{j'})[1+\gamma^2\mathcal{L}(\omega-\nu_1)]N_{10}, \tag{55}$$

$$M_{12}^r=\mathrm{Re}M_{12}=[(2\gamma_j/\gamma_{j'})\mathcal{L}_j(2\Delta)+(\gamma/\gamma_j')\mathcal{L}(\omega-\bar\nu)]N_{10}+[\gamma_j\gamma-2\Delta(\omega-\nu_1)]\mathcal{L}_j(2\Delta)\mathcal{L}(\omega-\nu_1)N_{12}, \tag{56}$$

$$M_{12}'^r=\mathrm{Re}M_{12}'=[(2\gamma_{j'}/\gamma_j)\mathcal{L}_{j'}(2\Delta)+(\gamma/\gamma_j)\mathcal{L}(\omega-\bar\nu)]N_{10}+[\gamma_{j'}\gamma-2\Delta(\omega-\nu_1)]\mathcal{L}_{j'}(2\Delta)\mathcal{L}(\omega-\nu_1)N_{12}, \tag{57}$$

$$Q_1^i=\mathrm{Im}Q_1=-[2\gamma(\omega-\nu_1)\mathcal{L}(\omega-\nu_1)/\gamma_j\gamma_{j'}]N_{10}, \tag{58}$$

$$M_{12}^i=\mathrm{Im}M_{12}=-[(4\Delta/\gamma_{j'})\mathcal{L}_j(2\Delta)+\gamma_{j'}^{-1}(\omega-\bar\nu)\mathcal{L}(\omega-\bar\nu)]N_{10}-[2\Delta\gamma+\gamma_j(\omega-\nu_1)]\mathcal{L}_j(2\Delta)\mathcal{L}(\omega-\nu_1)N_{12}, \tag{59}$$

$$M_{12}'^i=\mathrm{Im}M_{12}'=-[(4\Delta/\gamma_j)\mathcal{L}_{j'}(2\Delta)+\gamma_j^{-1}(\omega-\bar\nu)\mathcal{L}(\omega-\bar\nu)]N_{10}-[2\Delta\gamma+\gamma_{j'}(\omega-\nu_1)]\mathcal{L}_{j'}(2\Delta)\mathcal{L}(\omega-\nu_1)N_{12}, \tag{60}$$

$$\mathcal{L}(\nu)=(\gamma^2+\nu^2)^{-1}, \tag{61}$$

and

$$\mathcal{L}_\alpha(\nu)=(\gamma_\alpha^2+\nu^2)^{-1}. \tag{62}$$

C_{1x} is obtained from Eq. (54) by changing the over-all algebraic sign, interchanging the superscripts r and i, and finally multiplying the sine terms by minus one. It can furthermore be shown that S_{2x} and C_{2x} are obtained from S_{1x} and C_{1x} by interchanging the subscripts 1 and 2, while the corresponding y components are obtained by interchanging y and x.

In order to construct the final quantities needed in the self-consistant-field equations, it is necessary to find the projections of $\mathbf{S}_1$ and $\mathbf{C}_1$ along $\mathbf{E}_1$ and of $\mathbf{S}_2$ and $\mathbf{C}_2$ along $\mathbf{E}_2$. It is furthermore convenient to express these projections in a form that is independent of the coordinate system used for the calculation. The steps required to obtain this result are outlined in Appendix I. It is interesting to note that the calculation indicated in this Appendix only leads to results that are independent of the coordinate system if the identity

$$A=B+C+D \tag{63}$$

holds. This sum rule, after it has been disclosed on these purely physical grounds, was subsequently confirmed by the proof given in Appendix II.

Using the results of Appendix I in conjunction with Eqs. (31) and (54) yields

$$\begin{aligned}\mathbf{S}_1\cdot\mathbf{e}_1=&-(E_1/\hbar Ku)[\sum_m|\mathcal{P}_{mm}|^2]\,\mathrm{Im}Z(\nu_1-\omega)N_{10}+E_1^3(\pi^{1/2}/16\hbar^3Ku)Q_1^r[B+C+D(1-r_1^2)^2/(1+r_1^2)^2]\\&+E_1E_2^2(\pi^{1/2}/16\hbar^3Ku)\{BM_{12}^r+CM_{12}'^r+(CM_{12}^r+BM_{12}'^r)[(1-r_1^2)(1-r_2^2)/(1+r_1^2)(1+r_2^2)\cos^2\eta\\&+(r_1+r_2)^2/(1+r_1^2)(1+r_2^2)]+D(M_{12}^r+M_{12}'^r)\left[\frac{(1-r_1^2)(1-r_2^2)}{(1+r_1^2)(1+r_2^2)}\cos^2\eta+\frac{(r_1-r_2)^2}{(1+r_1^2)(1+r_2^2)}\right]\Bigg\},\end{aligned}\tag{64}$$

where r_1 and r_2 are the ratios of the minor to the major axes for the elliptically polarized radiation at the frequencies ν_1 and ν_2, respectively, and η is the angle between the two major axes. An expression for $\mathbf{C}_1\cdot\mathbf{e}_1$ can be obtained from Eq. (64) by changing the r superscripts to i's, and the corresponding quantities for frequency ν_2 can be obtained by interchanging the subscripts 2 and 1.

Inserting, in turn, the expressions for $\mathbf{S}_1\cdot\mathbf{e}_1$ and $\mathbf{S}_2\cdot\mathbf{e}_2$ obtained from Eq. (64) into Eq. (7) results, after a rearrangement of terms, in the amplitude-determining equations.

$$\dot{E}_1=\alpha_1E_1-\beta_1E_1^3-\theta_{12}E_1E_2^2 \tag{65}$$

and

$$\dot{E}_2=\alpha_2E_2-\beta_2E_2^3-\theta_{21}E_2E_1^2, \tag{66}$$

where

$$\alpha_1=-\tfrac{1}{2}(\nu/Q_1)+\tfrac{1}{2}\nu N_{10}(1/\epsilon_0\hbar Ku)[\sum_m|\mathcal{P}_{mm}|^2]\mathrm{Im}Z(\nu_1-\omega), \tag{67}$$

$$\beta_1=(\nu\pi^{1/2}/32\epsilon_0\hbar^3Ku)Q_1{}^r[B+C+D(1-r_1{}^2)^2/(1+r_1{}^2)^2], \tag{68}$$

and

$$\theta_{12}=\frac{\nu\pi^{1/2}}{32\epsilon_0\hbar^3Ku}\Bigg\{BM_{12}{}^r+CM_{12}{}'^r+(CM_{12}{}^r+BM_{12}{}'^r)\left[\frac{(1-r_1{}^2)(1-r_2{}^2)}{(1+r_1{}^2)(1+r_2{}^2)}\cos^2\eta+\frac{(r_1+r_2)^2}{(1+r_1{}^2)(1+r_2{}^2)}\right]$$
$$+D(M_{12}{}^r+M_{12}{}'^r)\left[\frac{(1-r_1{}^2)(1-r_2{}^2)}{(1+r_1{}^2)(1+r_2{}^2)}\cos^2\eta+\frac{(r_1-r_2)^2}{(1+r_1{}^2)(1+r_2{}^2)}\right]\Bigg\}, \tag{69}$$

and α_2, β_2, and θ_{21} are obtained from the quantities above by interchanging the subscripts 1 and 2.

Finally substituting, in turn, the expressions for $\mathbf{C}_1\cdot\mathbf{e}_1$ and $\mathbf{C}_2\cdot\mathbf{e}_2$ obtained from Eq. (64) into Eq. (6) yields the frequency-determining equations

$$\nu_1+\dot{\phi}_1=\Omega_1+\sigma_1+\rho_1E_1^2+\tau_{12}E_{12}^2 \tag{70}$$

and

$$\nu_2+\dot{\phi}_2=\Omega_2+\sigma_2+\rho_2E_2^2+\tau_{21}E_1^2, \tag{71}$$

where

$$\sigma_1=\tfrac{1}{2}\nu N_{10}(\epsilon_0\hbar Ku)^{-1}[\sum_m|\mathcal{P}_{mm}|^2]\mathrm{Re}Z(\nu_1-\omega), \tag{72}$$

and exchanging the superscripts r and i in Eqs. (68) and (69) gives the proper expressions for $-\rho$ and $-\tau_{12}$ respectively. These expressions are converted to the appropriate quantities for use in Eq. (71) by interchanging the subscripts 1 and 2.

V. DISCUSSION OF THE RESULTS

The amplitude- and frequency-determining expressions given above were obtained under the assumption, made in Sec. II, that the resultant fields are parallel to the polarization eigenstates of the passive cavity. We must now determine the realm of validity of this assumption and its effect upon our results. An examination of Eqs. (31) and (32), indicates that the first-order contribution to the polarization is parallel to the inducing field but that the third-order contribution is not. In the limit of small third-order effects, however, the direction of the total polarization approaches that of the first-order contribution, and our assumption is valid. Our assumption will also be valid for somewhat larger third-order effects if the frequency splitting between two modes of the same order number is large compared to the cavity width. In this case, the components of $\mathbf{E}_1$ and $\mathbf{E}_2$ perpendicular to $\mathbf{e}_1$ and $\mathbf{e}_2$ will be strongly attenuated by the cavity.

Since the errors introduced in the evaluation of the β's, θ's, and cavity losses are proportional to the cosine of the angle between the actual and assumed polarization directions, we would expect Eqs. (65) and (66) to provide a fairly accurate description of the oscillation amplitudes in the cases discussed above. Care must be exercised, however, in the use of the frequency-determining expressions, Eqs. (70) and (71), since a change in the states of polarization will result in a shift in the frequencies of the cavity resonances. This shift can often be comparable to the pulling and pushing effects caused by the dispersion of the medium. A discussion of these effects will thus be deferred pending a more detailed analysis of relationship between the present work and the cavity calculation.[2]

Before proceeding to a detailed treatment of the consequences of our results, it is useful to discuss some of the general features. We will thus examine the characteristics of the parameter θ_{12} [Eq. (69)] which expresses the strength of the third-order interaction between the two modes. The first two terms in Eq. (69) are independent of the polarization states of the fields, while the bracketed quantity in the third term, as one might expect, can be shown to be proportional to their scaler product. The polarization dependence of the last term, however, has no simple geometrical interpretation. This term, along with the corresponding term in β, as we will see below, leads to a number of rather unexpected predictions concerning laser performance. An analysis of the relatiohip betweensn the second- and third-order expressions for the density matrix components, Eqs. (25) and (26), reveals that the development of this last term involves, in second order, a coherent mixing of pairs of degenerate m states. As a result, third-order contributions to the polarization $\mathbf{P}_{mm}$ arise, or example, from successive excitation of the transitions $j,m\rightarrow j',m+1$; $j',m+1\rightarrow j,m+1$; and $j,m+1\rightarrow j',m$. The fact that these contributions do not in general have the same phase as the direct contributions to $\mathbf{P}_{mm}$, gives rise to the unique polarization dependence in Eq. (69). Portions of the terms involving B and C are also attributable to a coherent mixing of states. These contributions, however, involve the excitation of only two distinct transitions, and as a result, have the same phase as the direct contributions.

The discussions below will be primarily concerned with the values of the third-order interaction parameters. These parameters are related to the steady-state field intensities by the expressions

$$E_1^2=(\beta_2\alpha_1-\theta_{12}\alpha_2)/(\beta_1\beta_2-\theta_{12}\theta_{21}) \qquad (73)$$

and

$$E_2^2=(\beta_1\alpha_2-\theta_{21}\alpha_1)/(\beta_1\beta_2-\theta_{12}\theta_{21}) \qquad (74)$$

and enter into the condition for simultaneous oscillation of the two modes,

$$\theta_{12}\theta_{21}/\beta_1\beta_2\leqslant 1. \qquad (75)$$

In our discussions, the effects of atomic degeneracy and of the state of polarization will be illustrated by considering a number of special cases.

A. One Oscillating Mode

The dependence of the third-order polarization on the state of polarization of the inducing field has an important effect on laser output characteristics even when only one mode is oscillating. In this case, the steady-state intensity is given by

$$E^2=\alpha/\beta, \qquad (76)$$

where α and β are given by Eqs. (67) and (68). For convenience we will write

$$\beta_1=kQ_1^r[B+C+D(1-r_1^2)^2/(1+r_1^2)^2], \qquad (77)$$

where

$$k=\nu\pi^{1/2}/32\epsilon_0\hbar^3. \qquad (78)$$

It can be shown by use of the expressions for the matrix elements $\mathcal{P}_{mn}$ given in Appendix II, that if $j'=j\pm1$ the quantities B and C are always positive and D is always negative. Thus, for a given excitation rate, the laser output will be greatest for linear polarization ($r_1=0$ and $\beta_1=kQ_1^r[B+C+D]$) and least for circular polarization ($r_1=1$ and $\beta_1=kQ_1^r[B+C]$). This implies that, in the absence of cavity anisotropy, a single-mode laser with $j'=j\pm1$ will tend to oscillate in a linear state of polarization. This tendency does not exist if either the upper or lower atomic state is spherically symmetric ($j=0$). In these cases B and D are zero. If $j'=j$, the tendency is reversed.

B. Two Modes of Like Polarizations

In this case $r_1=r_2$, and $\eta=0$, so that Eq. (69) becomes

$$\theta_{12}=k(M_{12}^r+M_{12}'^r)\times[B+C+D(1-r_1^2)^2/(1+r_1^2)^2]. \qquad (79)$$

Equations (77) and (79) differ from Lamb's expressions for β_1 and θ_{12} only by the common factor $[B+C+D(1-r_1^2)^2/(1+r_1^2)^2]$. The coupling parameter $\theta_{12}\theta_{21}/\beta_1\beta_2$ is thus the same as that obtained by using Lamb's expressions. For mode splittings which are small compared to γ, the coupling parameter is greater than one (strong coupling). If we neglect the terms involving the proximity of the modes to the line center, the coupling parameter is four for zero splitting and one (critical coupling) for $2\Delta=(\gamma_j\gamma_{j'})^{1/2}$.

C. Orthogonally Polarized Modes

If we assume modes with opposite senses of rotation and perpendicular major axes, we have $r_2=-r_1$ and $\eta=\pi/2$, so that Eq. (69) becomes

$$\theta_{12}=k[BM_{12}^r+CM_{12}'^r+D(M_{12}^r+M_{12}'^r)(4r_1^2/(1+r_1^2)^2)]. \qquad (80)$$

If we again neglect terms involving proximity to the line center, Eq. (80), in the limit of zero splitting, becomes

$$\theta_{12}=kQ^r[B+C+2D(4r_1^2/(1+r_1^2)^2)]. \qquad (81)$$

Comparing Eqs. (77) and (81), we find that for orthogonal polarizations the value of the coupling parameter will depend upon the eccentricity of the polarization state, generally being greater than one at zero splitting for linear polarization and less than one for circular polarization when $\Delta j=\pm1$. For a transition involving a level with zero angular momentum, the coupling parameter for orthogonal modes is always one for zero splitting so that weak coupling occurs for any finite splitting. In this case Eqs. (77) and (80) reduce to

$$\beta_1=kCQ_1^r, \qquad (82)$$

and

$$\theta_{12}=kCM_{12}'^r. \qquad (83)$$

These last results are consistant with those obtained by Fork and Sargent[4] for circularly polarized oscillations and a transition between states with $j=1$ and $j'=0$. In making the comparison, it should be noted that, for mode splittings which are small compared to the separation of successive cavity resonances of the same polarization, N_{12} is approximately equal to N_{10}.

D. Modes Having Like Eccentricities and Opposite Senses of Rotation

Letting $r_2=-r_1$ but allowing η to vary, we have

$$\theta_{12}=k\left\{BM_{12}^r+CM_{12}'^r+(CM_{12}^r+BM_{12}'^r)\times\frac{(1-r_1^2)^2}{(1+r_1^2)^2}\cos^2\eta+D(M_{12}^r+M_{12}'^r)\times\left[\frac{(1-r_1^2)^2}{(1+r_1^2)^2}\cos^2\eta+\frac{4r_1^2}{(1+r_1^2)^2}\right]\right\}. \qquad (84)$$

This expression, the corresponding expression for θ_{21}, and the expressions for β_1 and β_2 obtained from Eq. (77) can be used in conjunction with the condition for

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

critical coupling, $\theta_{12}\theta_{21}/\beta_1\beta_2=1$, to provide an implicit relationship between the polarization and frequency characteristics of the laser output. It is clear from the forms of Eqs. (77) and (84) that this relationship could, in theory, be used to extract information about the decay constants associated with practical laser systems through measurements of laser output characteristics near critical coupling.

One of the most easily measured quantities associated with the polarization characteristics of two-mode laser output is the scalar product of the polarization vectors. An expression can be given explicitly relating this quantity to the frequency-dependent terms for the condition of critical coupling, if we take $\theta_{12}=\theta_{21}$, $\beta_1=\beta_2$, and neglect the terms in Eqs. (77) and (84) which involve D. The latter approximation is valid if either j or j' is zero and becomes steadily worse for larger values of j. If $j=1$ and $j'=2$, for example, $D\simeq\frac{1}{4}(B+C)$. We obtain

$$\left[\frac{(1-r_1^2)}{(1+r_1^2)}\right]\cos^2\eta=\frac{B(Q_1{}^r-M_{12}{}^r)+C(Q_1{}^r-M_{12}'{}^r)}{CM_{12}{}^r+BM_{12}'{}^r}. \quad (85)$$

This expression is in general agreement with measurements made in our laboratory of the polarization overlap and frequency splitting at critical coupling for a 3.5-μ He-Xe laser.

E. Combination Tones

An examination of the general expression for the third-order polarization, Eq. (32), reveals that, for two-mode operation, contributions to the polarization exist at the frequencies $2\nu_1-\nu_2$ and $2\nu_2-\nu_1$. Although the form assumed in Sec. II for the field in the cavity neglects the possibility of oscillations at these frequencies, it is clear that such combination tone oscillations can exist with appreciable strengths if the cavity losses at their frequencies are sufficiently small. This is indeed the case if the frequency splitting introduced by the cavity anisotropies is small compared to the cavity width. An interesting additional result of the theory is that the combination tones can exist even when the primary oscillations are orthogonal. This result is a direct consequence of the coherent mixing of degenerate m states discussed earlier.

We have previously reported the observation of combination tones in an anisotropic laser.[5] In these experiments, we were able to observe up to eight frequency components associated with a given pair of primary oscillations by using a high-gain laser with a low cavity Q.

VI. SUMMARY

We have developed above an optical-maser theory appropriate for systems involving degenerate atomic energy levels and arbitrarily polarized optical fields. In order to treat the case of a laser utilizing a generalized anisotropic cavity, we first derived self-consistent-field equations, allowing for the simultaneous existence of oscillations belonging to each of two distinct states of polarization. This derivation was performed under the simplifying assumption that the polarization states of the fields are completely determined by the passive cavity. Using a density-matrix formulation similar to that used by Lamb,[1] we next derived expressions for the first- and third-order contributions to the induced atomic polarization for a general laser transition between states with angular momentum quantum numbers j and $j'=j$, $j\pm1$ and complete m degeneracy. Explicit allowance was made in this calculation for the existence of oscillating modes corresponding to different laser cavity lengths. After specializing the polarization expressions to the case of two modes corresponding to different cavity lengths and possibly having different polarizations, we next extracted the projections of their in-phase and quadrature parts on the inducing field. These quantities were finally substituted into the self-consistent-field equations to obtain expressions for the frequencies and amplitudes of oscillation.

The third-order-interaction parameters which enter into our frequency and amplitude-determining expressions were found to depend in a fairly complicated way on the degree of atomic degeneracy and on the polarization states of the oscillations. The complicated nature of this dependence arises from terms in the third-order polarization which involve a coherent mixing of the degenerate states and leads to a number of interesting predictions regarding laser operation. In the case of single-mode operation, for example, it was found that if $\Delta j=\pm1$ third-order saturation is least for plane-polarized light. This indicates that in the absence of cavity anisotropies a single-mode laser will be plane-polarized.

For two-mode operation, particular attention was given to the condition for simultaneous oscillation. If the two polarization states are orthogonal, and $\Delta j=\pm1$, the value of the coupling parameter at zero splitting is generally less than one for circular polarization and greater than one for plane polarization. It is always equal to one for zero splitting if $j=0$ for either of the atomic levels. For two-mode operation with like polarizations, on the other hand, the competition parameter is unaffected by the atomic degeneracy or the state of polarization and is thus the same as that given by Lamb.

A number of topics mentioned in our discussions are appropriate for further study. One of the most interesting of these involves the possibility of using the relationship between frequency splitting and the polarization parameters at critical coupling to determine the values of the decay constants for the upper and lower atomic levels. It might also be quite interesting to investigate further the characteristics of the combination tones generated by neighboring oscillations and the relationship between polarization pulling by the laser medium and the output-frequency characteristics.

[5] Matthew B. White and Walter M. Doyle, Bull. Am. Phys. Soc. 10, 607 (1965).

APPENDIX I

In this Appendix, we trace the steps necessary to form the projections of the in-phase and quadrature components of the polarization on the inducing field. We first assume S_{1x}, for example, to be of the form

$$S_{1x}=a_{1x}E_{1x}+b_{11xx}E_{1x}^3+b_{11xy}E_{1x}E_{1y}^2+c_{12xx}E_{1x}E_{2x}^2 +c_{12xy}E_{1x}E_{2y}^2+c_{212xyy}E_{2x}E_{1y}E_{2y}, \quad (I.1)$$

with a similar form for S_{1y}. If we take $\mathbf{e}_1=\mathbf{E}_1/E_1$ where $E_1=|\mathbf{E}_1|$, we have

$$\mathbf{S}_1\cdot\mathbf{e}_1=|\mathbf{E}_1|^{-1}(S_{1x}E_{1x}+S_{1y}E_{1y}) \quad (I.2)$$

which can be written

$$\mathbf{S}_1\cdot\mathbf{e}_1=A_1E_1+B_1E_1^3+C_{12}E_1E_2^2 \quad (I.3)$$

with

$$A_1=(1+R_1^2)^{-1}(a_{1x}+a_{1y}R_1^2), \quad (I.4)$$

$$B_1=(1+R_1^2)^{-2}[b_{1x}+(b_{11xy}+b_{11yx})R_1^2+b_{1y}R_1^4], \quad (I.5)$$

and

$$C_{12}=[(1+R_1^2)(1+R_2^2)]^{-1}[c_{12xx}+c_{12xy}R_2^2 +(c_{212xyy}+C_{212yxx})R_1R_2 +c_{12yy}R_1^2R_2^2+c_{12yx}R_1^2], \quad (I.6)$$

where $R_1=E_{1y}/E_{1x}$ and $R_2=E_{2y}/E_{2x}$, and for the problem of interest, $a_{1x}=a_{1y}$, $b_{1x}=b_{1y}$, $b_{11xy}=b_{11yx}$, $c_{12xx}=c_{12yy}$, and $c_{12xy}=c_{12yx}$.

The dependence of Eq. (I.3) on the coordinate system can be removed by expressing R_1, R_2, and the phase factors which appear in the constants in terms of the eccentricities and relative orientations of the two elliptical states of polarization. We take α_1 and α_2 to be the angles which the two major axes make with the y axis, and r_1 and r_2 to be the ratios of the minor to major axes. These ratios are taken to be positive for clockwise rotation of the field and negative for counterclockwise rotation. It is sufficient to evaluate the factors $\cos 2\psi_1$, $\cos(\psi_1-\psi_2)$, and $\cos(\psi_1+\psi_2)$ which appear in b_{11xy}, c_{12xy}, and $c_{212xyy}+c_{212yxx}$. We find, for the last of these quantities,

$$\cos(\psi_1+\psi_2)=\frac{(1+R_1^2)(1+R_2^2)}{R_1R_2(1+r_1^2)(1+r_2^2)}[(1-r_1^2)(1-r_2^2) \times\sin\alpha_1\sin\alpha_2\cos\alpha_1\cos\alpha_2+r_1r_2]. \quad (I.7)$$

Cosine $2\psi_1$ can be obtained from this expression by setting $r_2=r_1$ and $\alpha_2=\alpha_1$, and $\cos(\psi_1-\psi_2)$ can be obtained by reversing the sign of r_1r_2. When these quantities are inserted into Eq. (I.3), and it is assumed that $A=B+C+D$ where A, B, C, and D are defined by Eqs. (40) through (43), the factors involving R_1 and R_2 cancel, and we obtain Eq. (64) where $\eta=\alpha_1-\alpha_2$. The sum rule assumed above is proven in the following Appendix.

APPENDIX II

In order to remove the coordinate system dependence in the preceding Appendix, it was necessary to assume the atomic sum rule $A=B+C+D$, where

$$A=\sum_m|\mathcal{P}_{mm}|^4, \quad (II.1)$$

$$B=\sum_m\{|\mathcal{P}_{m+1,m}|^2+|\mathcal{P}_{m-1,m}|^2\}|\mathcal{P}_{mm}|^2, \quad (II.2)$$

$$C=\sum_m\{|\mathcal{P}_{m,m-1}|^2+|\mathcal{P}_{m,m+1}|^2\}|\mathcal{P}_{mm}|^2, \quad (II.3)$$

and

$$D=\sum_m \mathcal{P}_{mm}\{\mathcal{P}_{m,m-1}\mathcal{P}_{m-1,m-1}\mathcal{P}_{m-1,m} +\mathcal{P}_{m,m+1}\mathcal{P}_{m+1m+1}\mathcal{P}_{m+1,m}\}. \quad (II.4)$$

We now prove this identity for the transition $j\rightarrow j+1$. If R is the reduced matrix element of the polarization operator between the states j and $j+1$, three of the matrix elements above become[6]

$$\mathcal{P}_{mm}=R[(j-m+1)(j+m+1)]^{1/2}, \quad (II.5)$$

$$\mathcal{P}_{m,m+1}=-\tfrac{1}{2}iR[(j-m+1)(j+m+1)]^{1/2}, \quad (II.6)$$

and

$$\mathcal{P}_{m,m-1}=-\tfrac{1}{2}iR[(j-m+1)(j-m+2)]^{1/2}. \quad (II.7)$$

The other matrix elements may be obtained by letting m become $m-1$ or $m+1$ in these expressions.

Carrying out the indicated algebraic operations, we find

$$B+C+D-A=\tfrac{1}{2}\sum_m[(j+1)^2-m^2] \times[5m^2-(j+1)^2+1]. \quad (II.8)$$

The values of the summations appearing in this expression are given by[7]

$$\sum_m m^2=\tfrac{1}{3}j(2j^2+3j+1), \quad (II.9)$$

$$\sum_m m^4=(1/15)j(6j^4+15j^3+j^2-1), \quad (II.10)$$

and

$$\sum_m k=(2j+1)k, \quad (II.11)$$

where k is a constant. Substitution of these quantities into Eq. (II.8) yields the desired result:

$$B+C+D-A=0.$$

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[6] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, New York, 1957).

[7] H. B. Dwight, *Tables of Integrals and other Mathematical Data* (The Macmillan Company, New York, 1947).