Theory of Magnetic Effects in Optical Maser Amplifiers and Oscillators*

C. V. HEER AND R. D. GRAFT†
Department of Physics, Ohio State University, Columbus, Ohio
(Received 21 June 1965)

A theoretical model is developed for the behavior of an optical maser medium in a static magnetic field of arbitrary strength in the Z direction and an electromagnetic field which is composed of traveling waves with both states of polarization and with both $+z$ and $-z$ directions. An integral formulation of a truncated phenomenological density matrix is introduced to treat the degeneracy of the atomic energy levels within the natural width. This integral is an exact solution of the phenomenological differential equation for the density matrix which was introduced by Wilcox and Lamb. The inherent degeneracy in the electromagnetic field is included by treating the field in a classical manner. The integral formulation is iterated to yield the first- and third-order contributions to the electric polarization vector $\mathbf{P}$ in a spatial element $d\mathbf{r}$, and the electric field and the electric polarization are made self-consistent in this spatial element by using coefficients in Maxwell's equations which are slowly varying functions of position and time. Atomic motion, hyperfine, isotope, and other line-broadening effects are included in the polarization vector $\mathbf{P}$. The general linear-amplifier, linear-oscillator, and closed-path-oscillator or photon-rate-gyroscope problems are formulated and in the simplest case require the analysis of eight nonlinear equations. These reduce to four equations for the linear oscillator with an axial magnetic field, and these examples are discussed in detail and compared with experiment. An anomalous situation arises for the $j_a=j_b$ or $F_a=F_b$ linear amplifiers or linear-oscillator masers, and the development indicates that these systems are stable near line center only for either right or left circular polarization in zero or weak magnetic fields. A magnetic field "dip" near line center is discussed. Special examples which require four or fewer nonlinear equations are discussed.

1. INTRODUCTION

EXPERIMENTAL investigations of gas optical masers, maser amplifiers, and related research on the atomic systems themselves require a more general expression for the macroscopic polarization than the expression introduced by Lamb[1] or the expressions introduced by Bloembergen and Shen.[2] Their discussions are limited to transitions in atoms with two nondegenerate energy levels and linearly polarized radiation. It is the purpose of this paper to extend the theory to real atoms which have states of angular momenta, and therefore, energy levels which are degenerate in the absence of a magnetic field and which in the presence of weak magnetic fields may lie within the natural width of the line. Some aspects of this problem have been considered by Tang and Statz[3] and by Culshaw and Kannelaud.[4] As in the previous papers the radiation field is treated in a classical manner, but the electric field which is perturbing an atom is now permitted to have arbitrary polarization and direction. In order to obtain the response of these atoms to the electric field or the macroscopic electric polarization of the gas in the region which is nonlinear in the electric field, an integral formulation of the density matrix is introduced. Since the density matrix is truncated to the energy levels of interest, damping and excitation are introduced in a manner similar to that used by Lamb and Wilcox[5] in their differential-equation formulation. This integral formulation is particularly convenient for treating degenerate systems or for determining the effect on the observable response of the system as the degeneracy is removed by a magnetic field. Some useful features of this integral formulation are shown in this paper, but it may be expected to be useful for other problems in which phenomenological differential equations of the Block[6] type have proved useful in the past.

Many of the interesting features in the third-order contribution to the macroscopic polarization are contained in a theory which includes electromagnetic waves traveling in the $+z$ and the $-z$ directions, of arbitrary polarization, and which are interacting with atoms in a static magnetic field in the Z direction. Explicit expressions are derived for this specific problem for the first-order polarization which is linear in the field and for the third order polarization which is nonlinear in the electric field. These expressions may be used to analyze a traveling-wave closed-path oscillator such as used in the photon-rate gyroscope,[7,8] the linear oscillator[9] discussed by Lamb, a linear oscillator with polarizing windows[10] and with the maser medium in a magnetic field, an amplifier with polarizing windows and again with the maser medium in a magnetic field, investigations of the natural width of the levels, etc. Second- and fourth-

* Supported in part by National Aeronautics and Space Agency Grant NsG-552.
† General Telephone Electronics Laboratory Predoctoral Fellow. Present address: Shell Chemical Co., Woodbury, New Jersey.
[1] W. E. Lamb, Jr., Phys. Rev. **134**, A1429 (1964).
[2] N. Bloembergen and Y. R. Shen, Phys. Rev. **133**, A37 (1964).
[3] C. L. Tang and H. Statz, Phys. Rev. **128**, 1013 (1962).
[4] W. Culshaw and J. Kannelaud, Phys. Rev. **133**, A691 (1964).
[5] L. R. Wilcox and W. E. Lamb, Jr., Phys. Rev. **119**, 1915 (1960).
[6] For general references, see A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon Press, Oxford, England, 1961).
[7] C. V. Heer, *Proceedings of the Third International Conference on Quantum Electronics* (Dunod Cie., Paris, and Columbia University Press, New York, 1963), and Phys. Rev. **134**, A799 (1964).
[8] W. Macek and D. Davis, Appl. Phys. Letters **2**, 67 (1963).
[9] A. Javan, W. R. Bennett, Jr., and D. R. Herriott, Phys. Rev. Letters **6**, 106 (1961).
[10] W. W. Rigrod, H. Kogelnik, D. J. Brangaccio, and D. R. Herrott, J. Appl. Phys. **33**, 743 (1962).

order tensors are introduced to relate the electric polarization to the electric field. If these are used with Maxwell's equations an index of refraction or accumulated phase becomes available and the type analysis which was used by Bennett[11] for maser oscillators may be used.

2. RESPONSE OF AN ATOM TO A PERTURBATION $V(t)$

The energy levels of a typical gas atom are shown in Fig. 1. Excitation to the sets of levels labeled $j_a m_a$ and $j_b m_b$ can occur because of collision with electrons, collision with photons of the trapped resonance radiation, or other excitation processes.[12] A perturbation $V(t)$ with matrix elements between the a-b levels induces an atomic polarization. In the presence of weak magnetic fields the separation of the energy levels of the m_a and m_b sets may be less than the natural line width. The perturbation $V(t)$ may include radiation of both states of polarization and radiation traveling in the $+z$ direction and $-z$ direction. Hence, a theory is needed which includes in its framework techniques for handling degeneracies in the atomic states and degeneracies in the radiation field. The density matrix provides a suitable framework. A density matrix for the atom plus radiation field is not used. The two are regarded as independent and the interaction of an atom with a perturbing field $V(t)$ is considered. Only the density matrix for the atom is needed in this approximation. The radiation field is expressed in a classical manner in terms of the field vectors **E** and **B** and in a certain sense includes the density-matrix approach for the radiation field since the number of photons in a cell of phase space is large.

If the Hamiltonian for an atom is H, then the development of the density matrix ρ in time is given by

$$i\hbar\dot{\rho}=H\rho-\rho H. \tag{1}$$

Since, in the problem under consideration, the observable effects are related to the a-b levels through the perturbation $V(t)$, it is convenient to truncate the density matrix to these levels. The remainder of the density matrix and the external perturbing terms in the Hamiltonian are included in a phenomenological manner. The phenomenological or psuedo-Boltzmann differential equation for the density matrix introduced by Lamb and Wilcox[5] is quite convenient and is suitable for treating many aspects of the problem under consideration.

$$i\hbar\dot{\rho}=(H_0-i\hbar\Gamma/2)\rho -\rho(H_0+i\hbar\Gamma/2)+i\hbar\lambda+(V\rho-\rho V); \tag{2}$$

ρ is the truncated part of the density matrix and has elements which are related to the a-b sets of states. H_0 is the Hamiltonian of the free atom and is independent of time. The anticommutator $\Gamma\rho+\rho\Gamma$ introduces damping

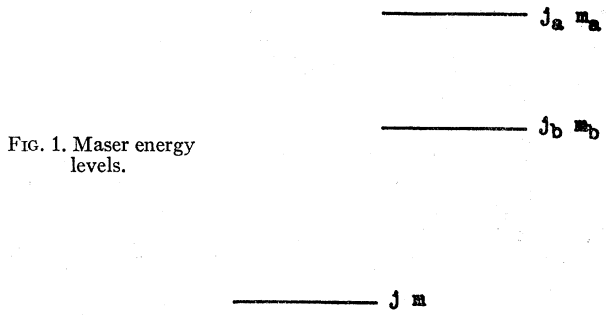

FIG. 1. Maser energy levels.

for the elements of the truncated density matrix and λ is the inhomogeneous excitation for these elements. Spontaneous emission from the excited levels is included in the damping term Γ. $V(t)$ contains the explicit time dependence.

If H_0, Γ, and λ commute, the phenomenological differential equation for ρ has an exact integral formulation,

$$\rho(t)=\lambda\Gamma^{-1}+(i\hbar)^{-1}\int_0^\infty dsT^\dagger(s) \times[V(t-s)\rho(t-s)-\rho(t-s)V(t-s)]T(s), \tag{3}$$

where $T(s)=\exp[-(\frac{1}{2}\Gamma-i\hbar^{-1}H_0)s]$. This integral form may be iterated to yield a series expansion of the density matrix. Regarding the perturbation $V(t)$ as a first-order nondiagonal quantity, iteration yields

$$\rho=\sum\rho^{(n)}, \tag{4a}$$

$$\rho^{(0)}=\lambda\Gamma^{-1}, \tag{4b}$$

$$\rho^{(n)}(t)=(i\hbar)^{-1}\int_0^\infty dsT^\dagger(s) \times[V\rho^{(n-1)}-\rho^{(n-1)}V]_{(t-s)}T(s). \tag{4c}$$

Since the integral formulation is exact and lends itself to a perturbation treatment of degenerate and nondegenerate problems, it seems appropriate to examine the requirement that H_0, Γ, and λ commute in greater detail. In the representation in which H_0 is diagonal, Γ and λ must be diagonal matrices. In this representation λ is a measure of the rate at which an atom enters a new state per second and Γ is a measure of the rate at which atoms leave this state. In the absence of the perturbation $V(t)$, $\rho=\lambda\Gamma^{-1}$ is diagonal and expresses the probability of occupation of the diagonal elements of the truncated density matrix. The probability of this occupancy is established by the external excitation processes and perturbations causing relaxation. If these processes are of thermal origin, then by using a canonical ensemble for ρ it follows that $\rho=\rho(H_0)=\exp(A-H_0)/kT=\lambda\Gamma^{-1}$.

In the more complex dynamical systems under consideration λ and Γ are determined by the experimental arrangement and spontaneous emission, and the thermal aspects are of minor importance. The damping matrix Γ

[11] W. R. Bennett, Jr., Appl. Opt., Suppl. 1, 24 (1962).
[12] Reference 11 contains a partial list of references to these processes.

is diagonal in the representation in which H_0 is diagonal for spontaneous emission.[13] Collisions which completely destroy the state add additional diagonal components to Γ and collisions which create only diagonal components of ρ are described by a diagonal matrix λ in the representation in which H_0 is diagonal. Although all aspects of relaxation cannot be covered in a brief manner, it would seem that rather than introduce nondiagonal elements to λ and Γ it would be better to enlarge the truncated density matrix in order to introduce the perturbation causing the nondiagonal elements. Thus in Fig. 1 the creation of a linear combination of states $m_a=\pm 1$ by the absorption of linearly polarized photons might be best treated by including the ground state into the truncated density matrix. An integral formulation of Eq. (2) with a single restrictive condition on Γ is given in a note added in proof.

Equations (2) and (3) describe the response of a single atom to a sequence of random perturbations λ and Γ and to a well-defined perturbation $V(t)$. The effect of collisions which destroy the phase relationship with the perturbing field $V(t)$ and effects which are related to the statistical average over a group of atoms will be included in subsequent sections.

In this paper the zero-order density matrix is assumed of the form

$$\rho^{(0)}(j_a m_a, j_a m_a; t)=\lambda(j_a m_a)\Gamma^{-1}(j_a m_a)\rightarrow \lambda_a/\Gamma_a, \quad (5)$$

etc. for the b levels, since the rate of entry and the rate of exit is assumed to be independent of the magnetic quantum numbers m. Direct substitution into Eq. (4) yields $\rho^{(n)}$ to all orders. Excitation to states a or b by trapped resonance radiation is included in the inhomogeneous excitation term λ.

3. ELECTRIC-DIPOLE TRANSITIONS

In a system in which only the levels j_a and j_b are connected by the time-dependent perturbation $V(t)$, the interaction of primary importance is the electric-dipole interaction and is of the form

$$V(t)=-\mathfrak{P}\cdot\mathbf{E}=-\sum_M(-)^M\mathfrak{P}_M{}^{(1)}E_{-M}; \quad (6)$$

$\mathfrak{P}$ is the electric dipole operator and $\mathfrak{P}_M{}^{(1)}$ is the same operator written as a tensor operator[14] of order 1. $\mathbf{E}$ is the electric field as measured by the atoms and E_{-M} the components of the electric vector in the spherical basis. This basis is defined in the conventional manner as

$$\hat{e}_\pm=\mp 2^{-1/2}(\hat{e}_X\pm i\hat{e}_Y); \quad \hat{e}_0=\hat{e}_Z, \quad (7)$$

and a general vector $\mathbf{E}$ may be expanded as

$$\mathbf{E}=\sum_{M=0,\pm1}(-)^M E_M\hat{e}_{-M} \quad \text{and} \quad E_M=\hat{e}_M\cdot\mathbf{E}. \quad (8)$$

The matrix elements of $\mathfrak{P}_M{}^{(1)}$ are conveniently obtained by the use of the Wigner-Eckart theorem,[14]

$$(j_a m_a|\mathfrak{P}_M{}^{(1)}|j_b m_b)=(2j_a+1)^{-1/2}\times(j_a\|\mathfrak{P}^{(1)}\|j_b)(j_b 1 m_b M|j_a m_a). \quad (9)$$

The notation of Ref. 14 is used for the Clebsch-Gordan coefficients and the reduced matrix elements. The observable time-dependent properties of the atoms are included in the trace $\rho\mathfrak{P}$. For a zero-order density matrix of the form given by Eq. (5), the first- and third-order quantities are of the form

$$\mathrm{tr}\rho^{(1)}\mathfrak{P}=K_1\sum_{m_a m_b}(-)^M\hat{e}_M(j_b 1 m_b M|j_a m_a)^2 i\int_0^\infty ds\exp[-\Gamma_{ab}-i\omega(m_a m_b)]sE_{-M}+\text{c.c.} \quad (10)$$

$$\mathrm{tr}\rho^{(3)}\mathfrak{P}=-K_2\sum_{m_a m_a',m_b m_b'}(j_b 1 m_b' M'|j_a m_a)(j_b 1 m_b' M'|j_a m_a')(j_b 1 m_b M''|j_a m_a')(j_b 1 m_b M'''|j_a m_a)(-)^{M'+M'''}\hat{e}_{M'''}$$
$$\times i\int_0^\infty ds\,ds'\,ds''\exp(-\Gamma_{ab}-i\omega(m_a m_b))s\{\exp(-\Gamma_b-i\omega(m_b' m_b))s'$$
$$\times[\exp(-\Gamma_{ab}-i\omega(m_a' m_b))s''E_{-M}E_{M'}'E_{-M''}''+\exp(-\Gamma_{ab}-i\omega(m_b' m_a'))s''E_{-M}E_{M'}''E_{-M''}']$$
$$+\exp(-\Gamma_a-i\omega(m_a m_a'))s'[\exp(-\Gamma_{ab}-i\omega(m_b' m_a'))s''E_{-M}'E_{M'}''E_{-M''}$$
$$+\exp(-\Gamma_{ab}-i\omega(m_a m_b'))s''E_{-M}''E_{M'}'E_{-M''}']\}+\text{c.c.}, \quad (11)$$

where

$$K_1=(\lambda_b\Gamma_b^{-1}-\lambda_a\Gamma_a^{-1})(2j_a+1)^{-1}\hbar^{-1}|(j_a\|\mathfrak{P}^{(1)}\|j_b)|^2, \quad (12)$$

$$K_2=(\lambda_b\Gamma_b^{-1}-\lambda_a\Gamma_a^{-1})(2j_a+1)^{-2}\hbar^{-3}\{|(j_a\|\mathfrak{P}^{(1)}\|j_b)|^2\}^2, \quad (13)$$

and

$$E_M=E_M(t-s), \quad E_M'=E_M(t-s-s'), \quad E_M''=E_M(t-s-s'-s''), \quad (14)$$

and

$$\Gamma_{ab}=\tfrac{1}{2}(\Gamma_a+\Gamma_b).$$

[13] G. Breit and I. S. Lowen, Phys. Rev. **46**, 590 (1934).

[14] The notation of A. Messiah, *Quantum Mechanics* (North-Holland Publishing Company, Amsterdam, 1962), Vol. II, is used for the tensor operators, Clebsch-Gordan coefficients, the Wigner-Eckart theorem, and the reduced matrix elements.

Although these equations are valid for an arbitrary electric field $\mathbf{E}$, waves traveling in the $\pm\hat{e}_z$ direction are of primary interest in this paper. For this purpose an almost plane monochromatic wave expansion

$$\mathbf{E}(z,t;k,\omega)=[\mathbf{E}(k,\omega)\exp-i\omega t+\mathbf{E}(k,-\omega)\exp i\omega t]\exp ikz+\text{c.c.} \tag{15}$$

is introduced. The $\mathbf{E}(k,\omega)$ are regarded as slowly varying functions of time and position in this approximate expansion. The E_M in Eqs. (10) and (11) are components of a real electric vector. Introduction of complex vectors $\mathbf{E}(k,\omega)$ requires greater care and to avoid confusion some of their properties are given. The coefficient $\mathbf{E}(k,\omega)$ refers to the wave traveling to the right and $\mathbf{E}(k,-\omega)$ to the wave to the left. $\mathbf{E}$ is transverse and $\text{div}\mathbf{E}\approx 0$. For convenience the substitution

$$\mathbf{E}(k,\omega)=\mathbf{A}=(\text{coefficient of wave to right}), \tag{16a}$$

$$\mathbf{E}(k,-\omega)=\mathbf{B}=(\text{coefficient of wave to left}) \tag{16b}$$

will be used in the third-order expansion. Spherical basis vectors are used in the expansion and $\mathbf{E}$ is expanded as

$$\mathbf{E}(z,t;k,\omega)=\sum_{M=0,\pm1}(-)^M E_M(z,t;k,\omega)\hat{e}_{-M}=\sum_{p=\pm1}(-)^p E_p\hat{e}_{-p} \tag{17a}$$

and

$$\begin{aligned}E_M(z,t;k,\omega)&=\hat{e}_M\cdot\mathbf{E}(z,t;k,\omega)\\&=[E_M(k,\omega)\exp-i\omega t+E_M(k,-\omega)\exp i\omega t]\exp ikz\\&\quad+[E_M^\dagger(k,\omega)\exp i\omega t+E_M^\dagger(k,-\omega)\exp(-i\omega t)]\exp(-ikz),\end{aligned} \tag{17b}$$

where $E_M(k,\omega)=\hat{e}_M\cdot\mathbf{E}(k,\omega)$. The superscript $\dagger$ is used to avoid confusion in the use of complex vectors and it should be noted that for $\mathbf{A}$ and $\mathbf{B}$

$$E_M^\dagger(k,\omega)=A_M^\dagger=(\hat{e}_M\cdot\mathbf{A}^*)=(-)^M(A_{-M})^*. \tag{18}$$

The choice of coordinate systems is shown in Fig. 2. XYZ refers to the coordinate system in which the static magnetic field is in the Z direction and is the reference system for the axis of quantization or the wave functions $\psi(jm)$. Since the radiation travels in the $\pm z$ direction, an xyz system is used for the radiation and the electric vector lies in the x-y plane. The components of a vector $\mathbf{A}$ in the x, y, z coordinate system are related to its components in the XYZ system by the $D_{Mp}^{(1)}(\alpha,\beta,\gamma)$ matrix.[15] If α is the rotation about the Z axis, β about the intermediate y' axis, and γ about the z axis, then in the spherical basis notation

$$A_M=\sum_{p=\pm1}D_{Mp}^*(\alpha,\beta,\gamma)A_p. \tag{19}$$

A_p refers to the x, y, z system and $p=\pm1$ since the radiation is in the z direction. $p=+1$ denotes right circular and $p=-1$ denotes left circular polarization for a wave traveling in the $+z$ direction. A_M refers to the components in the XYZ system and $M=0,\pm1$ can then occur. Pertinent details of the transformation are given in Appendix I.

4. MACROSCOPIC ELECTRIC POLARIZATION

The macroscopic electric polarization and the density matrix are related by

$$\mathbf{P}(r,t)=N\,\text{tr}\rho(\mathbf{r},t)\mathfrak{P}=N\langle\text{tr}\rho(\mathbf{r},t;\mathbf{v})\mathfrak{P}\rangle, \tag{20}$$

where the $\langle\ \rangle$ indicate an average over the atomic velocity distribution in a small spatial element $d\mathbf{r}$. The development in the previous section gives $\rho(t)$ for atoms which experience a perturbation $V(t)$ as measured by the atoms. If an atom enters state $(j_a m_a)$ at position $\mathbf{r}_e$ at time t_e with velocity $\mathbf{v}$, it will contribute[1] at $\mathbf{r}$ at time t if

$$\mathbf{r}_e=\mathbf{r}-\mathbf{v}(t-t_e).$$

The electric field $\mathbf{E}(t')$ measured by the atoms during the interval $(t-t_e)$ is related to the radiation field by

$$\mathbf{E}(t')=\mathbf{E}(\mathbf{r}_e+\mathbf{v}(t'-t_e);t')=\mathbf{E}(\mathbf{r}-\mathbf{v}(t-t');t').$$

Since the atomic perturbation for an electric dipole is

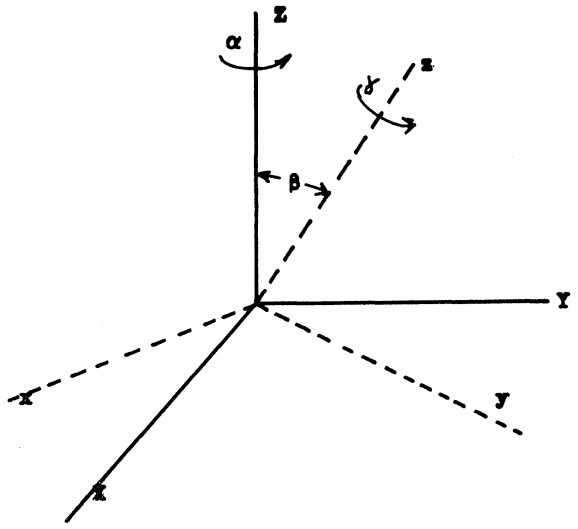

FIG. 2. Magnetic field $\mathfrak{B}$ is along Z and determines the axis of quantization. The radiation is along $\pm z$ with the electric vector in the xy plane. $\alpha\beta\gamma$ are the Euler angles.

[15] M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957).

of the form given by Eq. (6) the density matrix for the group of atoms with velocity $\mathbf{v}$ is

$$\rho(t)\rightarrow\rho(\mathbf{r},t;\mathbf{v}),\quad\text{with}\quad V(t-s)\rightarrow V(\mathbf{r}-\mathbf{v}s;t-s). \tag{21}$$

Only the motion in the direction of the traveling wave has a significant effect. If $W(v_z)$ is the probability that an atom enters state j with velocity v_z which is along the direction of the traveling wave, then $\lambda\rightarrow\lambda W(v_z)$ and

$$\rho(z,t)=\langle\rho(z,t;v_z)\rangle=\int_{-\infty}^{\infty}dv_zW(v_z)\rho(z,t;v_z). \tag{22}$$

For a Doppler-broadened line, the typical integral is of the form

$$\int_{-\infty}^{\infty}dv_zW(v_z)\exp\pm ikv_zs=\exp-\tfrac{1}{4}D^2s^2, \tag{23}$$

where

$$D^2=8\pi^2kT\nu^2/mc^2=\pi^2\Delta\nu_D{}^2/\ln 2, \tag{24}$$

and $\Delta\nu_D$ is the Doppler width. The Doppler-broadening integral

$$\begin{aligned}&Z(\omega-\omega_{ab},\Gamma_{ab},D)\\&=\left\langle iD\int_0^{\infty}ds\exp[-\Gamma_{ab}+i(\omega-\omega_{ab})\pm ikv_z]s\right\rangle\\&=iD\int_0^{\infty}ds\exp[-\Gamma_{ab}s+i(\omega-\omega_{ab})s-\tfrac{1}{4}D^2s^2]\end{aligned} \tag{25}$$

is of use in the first-order expansions.[16]

The macroscopic electric polarization is assumed to have a form similar to that used for the electric field,

$$\begin{aligned}\mathbf{P}(z,t;k,\omega)=&[\mathbf{P}(k,\omega)\exp(-i\omega t)+\mathbf{P}(k,-\omega)\exp i\omega t]\\&\times\exp ikz+\text{c.c.}+\text{higher order terms}.\end{aligned} \tag{26}$$

$P_M(k,\omega)$ coefficients are introduced in the same manner as used in Eq. (15) for the electric field and $P_M{}^{(1)}(k,\omega)$ is used to denote a typical term linear in the electric field and $P_M{}^{(3)}(k,\omega)$ a typical third-order term in the electric field, or

$$P_M(k,\omega)=P_M{}^{(1)}(k,\omega)+P_M{}^{(3)}(k,\omega)+\cdots. \tag{27}$$

The first- and third-order polarization coefficients are now given for this model.

First-Order Polarization Coefficients

Direct substitution into Eq. (10) and averaging over the velocity distribution yields the first order coefficients to the macroscopic polarization

$$\begin{aligned}&P_{-M}{}^{(1)}(k,\omega)\\&=ND^{-1}K_1\sum_{m_a,m_b}(j_b1m_bM\,|\,j_am_a)^2\\&\qquad\times Z(\omega-\omega(m_a,m_b),\Gamma_{ab},D)E_{-M}(k,\omega),\end{aligned} \tag{28a}$$

$$\begin{aligned}&P_M{}^{(1)}(k,-\omega)\\&=ND^{-1}K_1\sum_{m_a,m_b}(j_b1m_bM\,|\,j_am_a)^2\\&\qquad\times Z^*(\omega-\omega(m_a,m_b),\Gamma_{ab},D)E_M(k,-\omega),\end{aligned} \tag{28b}$$

and Z is defined by Eq. (25).

Third-Order Polarization Coefficients

Direct substitution in Eq. (11) and averaging over the velocity distribution yields the third-order coefficients to the macroscopic polarization.

$$\begin{aligned}P_{-M'''}{}^{(3)}(k,\omega)=&-iNK_2\sum_{m_am_a',m_bm_b'}(-)^{M'}(j_b1m_b'M\,|\,j_am_a)(j_b1m_bM'\,|\,j_am_a')(j_b1m_bM''\,|\,j_am_a')(j_b1m_bM'''\,|\,j_am_a)\\&\times\Big\langle\int_0^{\infty}dsds'ds''\exp[-\Gamma_{ab}-i(\omega(m_am_b)-\omega)]s\{\exp[-\Gamma_b-i\omega(m_b'm_b)]s'\\&\times[\exp[-\Gamma_{ab}-i(\omega(m_a'm_b)-\omega)]s''(A_{-M}A_{M'}{}^{\dagger}A_{-M''}G_1+A_{-M}B_{M'}B_{-M''}{}^{\dagger}G_2+B_{-M}{}^{\dagger}B_{M'}A_{-M''}G_3)\\&+\exp[-\Gamma_{ab}+i(\omega(m_a'm_b')-\omega)]s''(A_{-M}B_{M'}B_{-M''}{}^{\dagger}G_1+A_{-M}A_{M'}{}^{\dagger}A_{-M''}G_2+B_{-M}{}^{\dagger}B_{M'}A_{-M''}G_3)]\\&+\exp[-\Gamma_a-i\omega(m_am_a')]s'[\exp[-\Gamma_{ab}-i(\omega(m_am_b')-\omega]s''\\&\times(A_{-M}A_{M'}{}^{\dagger}A_{-M''}G_1+A_{-M}B_{M'}B_{-M'}{}^{\dagger}G_3+B_{-M}{}^{\dagger}B_{M'}A_{-M''}G_2)+\exp[-\Gamma_{ab}+i(\omega(m_a'm_b')-\omega)]s''\\&\times(A_{-M}B_{M'}B_{-M''}{}^{\dagger}G_3+A_{-M}A_{M'}{}^{\dagger}A_{-M''}G_2+B_{-M}{}^{\dagger}B_{M'}A_{-M''}G_1)]\}\Big\rangle.\end{aligned} \tag{29a}$$

$$P_{M'''}{}^{(3)}(k,-\omega)=[\text{change } i \text{ to } -i \text{ and } A_M \text{ to } B_{-M}, \text{ etc., in Eq. (29a)}]. \tag{29b}$$

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

TABLE I. Nonzero products of the four Clebsch-Gordan (C-G) coefficients contributing to $\chi(-M''', -M, M', -M'')$. The symbol

$$\{\text{C-G}\}=(j_b 1 m_b' M' \mid j_a m_a)(j_b 1 m_b' M' \mid j_a m_a')(j_b 1 m_b M'' \mid j_a m_a')(j_b 1 m_b M''' \mid j_a m_a);$$

and the C-G coefficients imply $-M'''=-M+M'-M''$; $m_a-m_a'=M'''-M''=M-M'$, $m_b-m_b'=M-M'''=M'-M''$, etc. The $\Sigma_{m_a}\{\text{C-G}\}$ has the following sum rules: (1) = ½[(9)+(7)+(18)] = ½[(8)+(6)+(19)]; (8) = (9) = ½[(2)+(4)+(15)+(17)] = ½[(3)+(5)+(14)+(16)]; (9) − (7) − (18) = (10) + (13) = (12) + (11); and (2) = (4) = (3) = (5); (6) = (7); (10) = (11) = (12) = (13); (14) = (15) = (16) = (17); (18) = (19). Each dash in column (3) denotes a nonzero Clebsch-Gordon coefficient between levels $(j_a m_a)$ and $(j_b m_b)$. Four products occur in the symbol {C-G}, and a vertical dash | implies the change in m is zero, while slanting dashes \ or / imply the change in m is ±1.

$\chi(-M''', -M, M', -M'')$		$R\Sigma_{m_a}\{\text{C-G}\}$							$R\Sigma_{m_a} m_a\{\text{C-G}\}$				
j_a or F_a		1	2	1	2	3	⅜	⅝	1	2	1	2	3
j_b or F_b		0	1	1	2	2	⅜	⅜	0	1	1	2	2
R		1	36	4	36	225	225	100	1	36	4	36	225
1 (0000)	||||	1	34	2	34	259	164	104	0	0	2	0	0
2 (00+−)	||/	1	33	1	13	288	68	98	0	15	−1	−17	196
3 (−−00)	||/	1	33	1	13	288	68	98	1	48	0	−4	434
4 (00−+)	\\\||	1	33	1	13	288	68	98	0	−15	1	17	−196
5 (++00)	\\\||	1	33	1	13	288	68	98	−1	−48	0	4	−434
6 (−−−+)	\\/	1	21	1	21	126	96	56	1	21	1	21	101
7 (+++−)	\\/	1	21	1	21	126	96	56	−1	−21	−1	−21	−101
8 (−−+−)	////	1	46	2	26	371	136	146	1	81	1	13	910
9 (++−+)	\\\\	1	46	2	26	371	136	146	−1	−81	−1	−13	−910
10 (0−0+)	|×|	0	12	0	−8	112	−28	42	0	6	0	−4	56
11 (−0−0)	|×|	0	12	0	−8	112	−28	42	0	6	0	−4	56
12 (+0+0)	|×|	0	12	0	−8	112	−28	42	0	−6	0	4	−56
13 (0+0−)	|×|	0	12	0	−8	112	−28	42	0	−6	0	4	−56
14 (−00−)	//||	0	13	1	13	133	68	48	0	9	1	17	140
15 (0−+0)	//||	0	13	1	13	133	68	48	0	9	1	17	140
16 (+00+)	||\\	0	13	1	13	133	68	48	0	−9	−1	−17	−140
17 (0+−0)	||\\	0	13	1	13	133	68	48	0	−9	−1	−17	−140
18 (+−++)	/\/\	0	1	1	21	21	96	6	0	0	0	0	0
19 (−+−−)	/\/\	0	1	1	21	21	96	6	0	0	0	0	0

Again it is useful to note that $A_M^\dagger=(-)^M(A_{-M})^*$. The G coefficients are of the form $\exp(\pm ikv_z s)$ and the average over the Doppler width is given by Eq. (23) as

$$\langle G(s)\rangle=\langle\exp(\pm ikv_z s)\rangle=\exp-\tfrac{1}{4}D^2s^2, \tag{30}$$

where

$$G_1=G(s+s''); \quad G_2=G(s-s''); \quad \text{and} \quad G_3=G(s+2s'+s'').$$

5. TENSOR FORMULATION FOR THE ELECTRIC SUSCEPTIBILITY

Equations (28) and (29) suggest a tensor formulation for the polarization in terms of the spherical basis functions. If M in Eq. (28) is specified, then the sum is limited to m_a or m_b by the Clebsch-Gordan (C-G) coefficient. If M''' is specified in Eq. (29) the sum over either m_a or m_b may be omitted. If the other values of M, M', M'' are specified, the sum is reduced to the sum over a single value of m, or

$$\sum_{m_a m_a', m_b m_b'} \cdots \longrightarrow \sum_{m_a M M' M''} \delta(M'''-M+M'-M'')\cdots. \tag{31}$$

The Kronecker δ indicates the constraint which is placed on the M indices by the Clebsch-Gordan coefficients and Table I indicates the 19 nonzero products of the C-G coefficients in terms of the M indices. If more convenient, m_b may be chosen as the summation index. The polarization may be written in tensor form as

$$\begin{aligned}\epsilon_0^{-1}P_{-M'''}(k,\omega)=&\chi(-M''', M'''; k,\omega)A_{-M'''}+\chi_a(-M''', -M, M', -M''; k,\omega)A_{-M}A_{M'}{}^\dagger A_{-M''}\\&+\chi_b(-M''', -M, M', -M''; k,\omega)A_{-M}B_{M'}B_{-M''}{}^\dagger\\&+\chi_c(-M''', -M, M', -M''; k,\omega)B_{-M}{}^\dagger B_{M'}A_{-M''}.\end{aligned} \tag{32a}$$

$$P_{M'''}(k,-\omega)=(\text{change } i \text{ to } -i \text{ in } \chi \text{ and } A_{-M} \text{ to } B_M, \text{ etc.}). \tag{32b}$$

Repeated indices are summed in these expressions. χ_b and χ_c are coefficients of the coupling between the $\pm z$ traveling waves.

A typical $\chi(-M''', -M'''; k,\omega)$ coefficient may be obtained by inserting the desired value of M''' in Eq. (28) and then summing over m_a or m_b. This second order tensor is diagonal in the M indices and χ and Z are related by

$$\epsilon_0\chi(-M''', -M'''; k, \omega) = ND^{-1}K_1 \sum_{m_a \text{ or } m_b} (j_b 1 m_b M''' | j_a m_a)^2 \times Z(\omega-\omega(m_a m_b); \Gamma_{ab}, D). \quad (33)$$

Since the Z functions are tabulated[16] $\chi(-M''', -M''')$ may be evaluated in principle for arbitrary values of the magnetic field and ω.

No such simple procedure is possible for the third-order terms. In general

$$\epsilon_0\chi_a(-M''', -M, M', -M''; k, \omega) = -iNK_2 \sum_{m_a} (\text{coefficient of } A_{-M}A_{M'}{}^{\dagger}A_{-M''} \text{ in Eq. (29a)}). \quad (34)$$

with similar expressions for χ_b and χ_c. For atoms at rest

$$G_1 = G_2 = G_3 = 1, \quad (35)$$

and the typical integrals in Eq. (29a) over s, s', s'' yield Lorentz line shapes or products of the type

$$[+i(\omega(m_a m_b)-\omega)+\Gamma_{ab}]^{-1}[i\omega(m_b' m_b)+\Gamma_b]^{-1} \times [i(\omega(m_a' m_b')-\omega)-\Gamma_{ab}]^{-1}. \quad (36)$$

Since integrals of this type are readily evaluated only one of the four products is given. A particular feature of two of the four integral products is the tendency for the effect of the frequency ω to become of reduced importance. This is more noticeable as the average over the velocities is made and in the extreme Doppler limit ω cancels out of two of the four repeated integrals. For large Doppler broadening the contributions of G_1 and G_3 are small compared to that of G_2, and the G_2 term tends to act as a δ function,[1] so that

$$\int_0^\infty ds ds'' f(s,s'')G(s-s'') \rightarrow 2\pi^{1/2}D^{-1} \times \int_0^\infty ds' f(s's'')\delta(s-s''). \quad (37)$$

Cancellation is apparent in the second and fourth integrals in Eq. (29a) and two of the products for extreme Doppler motion yield line shapes of the form

$$D^{-1}[i(\omega(m_a m_b)-\omega(m_a' m_b'))+2\Gamma_{ab}]^{-1} \times [i\omega(m_b' m_b)+\Gamma_b]^{-1}, \quad (38a)$$

while the remaining two integrals yield lines shapes of the form

$$D^{-1}[i(\omega(m_a m_b)+\omega(m_a' m_b)-2\omega)+2\Gamma_{ab}]^{-1} \times [i\omega(m_b' m_b)+\Gamma_b]^{-1}. \quad (38b)$$

Large Doppler broadening tends to cancel the coupling between $+z$ traveling waves and $-z$ traveling waves. Averaging over the atomic velocities tends to reduce this coupling and to enhance the cancellation of ω. The frequency dependence of the two integrals contributing to χ_a are of the form given by Eq. (38a); $i\omega(m_b' m_b)+\Gamma_b$ occurs in one integral and $i\omega(m_a m_a')+\Gamma_a$ in the other. The frequency dependence of χ_b is given by Eq. (38b) and χ_c differs in the m indices as indicated in Eq. (29a).

All other cases lie between these two extremes. Aronowitz[17] has considered the contribution of integrals of the type $\langle G_1\rangle$ and $\langle G_3\rangle$ in some detail.

6. MODIFICATIONS FOR HYPERFINE AND ISOTOPE EFFECTS

Zeeman Splitting of the Energy Levels

In the absence of hyperfine splitting, the splitting of the magnetic sublevels is given by

$$(j_a m_a | -\mathbf{M}\cdot\mathfrak{B} | j_a m_a) = [-(2j_a+1)^{-1/2}(j_a\|\mathfrak{M}^{(1)}\|j_a)]m_a\mathfrak{B} = \hbar\gamma_a m_a\mathfrak{B}, \quad (39)$$

where the reduced matrix element is replaced by γ_a. The energy difference between optical levels may then be expressed as

$$\omega(m_a m_b)-\omega_{ab} = (\gamma_a m_a - \gamma_b m_b)\mathfrak{B}. \quad (40)$$

Hyperfine and Magnetic-Field Splitting of Energy Levels

The expression for $P_M{}^{(1)}$ and $P_M{}^{(3)}$ may be generalized to atoms with hyperfine splitting by making the following changes in Eqs. (28) and (29). The Clebsch-Gordan coefficients and the reduced matrix elements are changed to

$$(j_b 1 m_b M | j_a m_a) \rightarrow (F_b 1 m_{F_b} M | F_a m_{F_a})$$
$$(2j_a+1)^{-1/2}(j_a\|\mathfrak{P}^{(1)}\|j_b) \rightarrow (2F_a+1)^{-1/2} \times (j_a I F_a\|\mathfrak{P}^{(1)}\|j_b I F_b). \quad (41)$$

The trace must now include the sum over the hyperfine levels and is of the form,

$$\sum_{m_a m_b} \rightarrow \sum_{F_a m_{F_a}} \sum_{F_b m_{F_b}}. \quad (42)$$

An expression for the energy-level separation

$$\omega(F_a m_{F_a}, F_b m_{F_b})-\omega_{ab} \quad (43)$$

is given in Appendix II along with other pertinent data. Although most atoms have nuclear spin and hyperfine splitting the notation $(j_a m_a)$ is used in most of this

[17] F. Aronowitz, Phys. Rev. 139, A635 (1965).

paper to avoid the double subscript m_{F_a}. The transformation from one notation to the other is readily made by the substitutions given above. Table I may be used by changing the j's to F's.

Isotopes

If more than one isotope is present, then

$$P=N\,\mathrm{tr}\rho\mathfrak{P}\rightarrow\sum_{\alpha}N_{\alpha}\,\mathrm{tr}\rho_{\alpha}\mathfrak{P}\,, \tag{44}$$

where N_α is the number of the αth isotope atoms per cubic meter and ρ_α is the density matrix for the αth isotope atoms. Since the isotopes will in general have differences in nuclear spin and mass, ρ_α will differ in the zero field splitting ω_{ab}, the hyperfine splitting, and in the F, m_F indices over which the trace is summed. In this same manner the electric susceptibility is an average over the individual susceptibilities for each isotope,

$$\chi(M''',M,M',M'')=\sum_{\alpha}(N_\alpha/N)\chi_\alpha(M''',M,M',M'')\,.$$

Weak and Strong Magnetic Fields

In zero magnetic field the polarization is independent of the choice of the axis of quantization and for the entries in Table I the integrands are independent of m_a, m_b, etc. In the presence of hyperfine splitting it is necessary to complete the sums over F_a and F_b and χ will depend on the amount of hyperfine separation in comparision with the Doppler width in the first-order terms and on the natural width in the third-order terms. In the absence of hyperfine splitting χ is pure imaginary in zero field at line center. χ is complex when ω is not at line center and is in general complex for atoms with hyperfine splitting. It is approximately correct to consider for

$$\chi=\chi'+i\chi''$$

that χ' causes a phse shift and χ'' effects the gain.

A magnetic field is weak if the splitting of the energy levels is small compared to the natural width of the line. The width of the natural line is dominant in the third-order terms and the Doppler width in first order terms. As the magnetic field is increased from a value of zero for an atom without hyperfine structure the term χ' will grow linearly with the field and interference effects between states of polarization can be expected near line center. This is no longer true for atoms away from line center or for atoms with hyperfine splitting and a linear dependence in the gain can occur. This linear dependence is proportional to the product of the Zeeman splitting with the deviation from line center or with the hyperfine splitting. Weak magnetic fields require a consideration of all three values of M in $\chi(M,M)$ and of all terms shown in Table I for the third-order term $\chi(M''',M,M',M'')$.

In strong magnetic fields, that is fields which yield a Zeeman splitting large compared to the natural width, the major contribution in third order arises from one of the terms of type 1, 8, or 9 in Table I. The frequency ω determines which term is most significant. ω also determines which of the three first order values of $\chi(MM)$ has the largest gain. Thus a strong magnetic field tends to create a two level problem, and which two levels depends on ω and on the polarization of the radiation.

7. ELECTROMAGNETIC FIELD EQUATIONS

The development in the previous section gives the electric polarization in a space element $d\mathbf{r}$ if the electric field is known. Maxwell's equations require the electric polarization and the electric field in the space element to be related by

$$-\mathrm{curl}\,\mathrm{curl}\mathbf{E}-c^{-2}\partial^2\mathbf{E}/\partial t^2=c^{-2}\epsilon_0^{-1}\partial^2\mathbf{P}/\partial t^2 \tag{45}$$

and div$\mathbf{E}$ is assumed approximately zero for the system under consideration. In the experiments under consideration a large number of photons are assumed to be traveling in the $\pm z$ directions and these almost plane waves have two states of polarization for the electric vector. $\hat{a}_x$ and $\hat{a}_y$ form one convenient expansion set for the polarization. Since a spherical basis set is used for the expansion of the atomic states, it is convenient to introduce a spherical basis set $\hat{a}_\pm=\mp 2^{-1/2}(\hat{a}_x\pm i\hat{a}_y)$ for the expansion set for the radiation field. Following the procedure indicated in Eqs. (15)–(17) the electric field is expanded in terms of the components

$$E_p(k,\omega)\,\exp i(kz-\omega t)\,, \tag{46}$$

where $p=\pm 1$ and $E_p(k,\omega)$ is regarded as a slowly varying function of t and z. $E_p(k,\omega)$ may also vary with the beam cross section in the manner suggested by Fox and Li[18] for a maser or in other suitable ways to limit the beam of radiation to a finite cross section.[19] Neglecting second derivatives in space and time, Eq. (45) may be written in the approximate form

$$2ikc^2\frac{\partial E_p(k,\omega)}{\partial z}+2i\omega\frac{\partial E_p(k,\omega)}{\partial t}$$
$$+(\omega^2-k^2c^2)E_p(k,\omega)=-\epsilon_0^{-1}\omega^2P_p(k,\omega)\,. \tag{47}$$

Equation (47) implies four equations since $p=\pm 1$ and changing ω to $-\omega$ yields new equations. Four additional equations are obtained by changing k to $-k$ and E_p to $E_p^\dagger$ or by noting that E_p is complex. These eight equations relate the electric field and the electric polarization in a volume element $d\mathbf{r}$ and the derived value of the polarization in terms of the field must be self-consistent with these equations.[1]

Unfortunately the polarization is given most conveniently in the XYZ coordinate system and the radia-

[18] A. G. Fox and T. Li, Bell System Tech. J. **40**, 453 (1961).
[19] S. A. Collins, Jr., Appl. Opt. **3**, 1263 (1964); P. O. Clark, J. Appl. Phys. **36**, 66 (1965).

tion field in the xyz system. The first- and third-order contribution to the polarization may be transformed by using the D_{Mp} coefficients given in Eq. (19). Thus

$$\epsilon_0^{-1}P_{-p'''}{}^{(1)}(k,\omega)=D(-M''',-p''')D^*(-M''',-p)\times\chi(-M''',-M''')A_{-p}=\chi(-p''',-p)A_{-p} \quad (48)$$

or the second-order tensor transforms as

$$\chi(-p''',-p)=D(-M''',-p''')D^*(-M''',-p)\times\chi(-M''',-M''';k,\omega). \quad (49)$$

It is convenient to have the index p in Eq. (47) denote two canonical states of polarization α, and these canonical states can be selected such that $\chi(\alpha'\alpha)$ is diagonal. This may be accomplished by using a transformation of the type

$$A_M=D^*(Mp)A_p=D^*(Mp)U^*(p\alpha)A_\alpha=S^*(M\alpha)A_\alpha, \quad (50)$$

where U and S are unitary matrices and $\alpha=1, 2$ is an index denoting the two canonical states of polarization. The transformation from P_M to P_α is given by Eq. (A5) in Appendix I. Equations (28a) and (29a) may be transformed directly by transforming P_M as

$$P_{\alpha'''}(k,\omega)=S(-M''',\alpha''')P_{-M'''}(k,\omega), \quad (51a)$$

and by noting that

$$A_{-M}A_{M'}{}^\dagger A_{-M''}=(-)^{M'}S^*(-M\alpha)S(-M'\alpha')\times S^*(-M''\alpha'')A_\alpha(A_{\alpha'})^*A_{\alpha''} \quad (51b)$$

etc. Equations (28) and (29) are not rewritten in terms of the α index since the substitution of Eqs. (51a) and (51b) into Eqs. (28) and (29) is readily made.

Using the approximation that $\omega\approx kc$, the equation of motion of the field in terms of the A's and B's is for either the p or α index,

$$2ik^{-1}\frac{\partial A_\alpha}{\partial z}+2i\omega^{-1}\frac{\partial A_\alpha}{\partial t}=-\epsilon_0^{-1}P_\alpha(k,\omega), \quad (52a)$$

$$2ik^{-1}\frac{\partial B_\alpha}{\partial z}-2i\omega^{-1}\frac{\partial B_\alpha}{\partial t}=-\epsilon_0^{-1}P_\alpha(k,-\omega), \quad (52b)$$

where P_α is given by Eq. (51) or (A5). The equation for $E_\alpha(k,-\omega)$ or B_α is obtained by changing A_α to B_α, ω to $-\omega$ and using $P_\alpha(k,-\omega)$. Again eight equations are implied, that is, for the real and imaginary parts of A_α and B_α. These equations are nonlinear and simple solutions will only occur for special cases. The equations are examined in the ensuing sections in the order of their complexity.

8. AMPLIFIER WITH AXIAL MAGNETIC FIELD

The simplest problem to consider is an amplifier with an axial magnetic field since D_{Mp} is a diagonal unit matrix and $M=p=\pm1$. In this initial example it is assumed that no reflected wave occurs and the coefficients of the traveling wave in the $-z$ direction are zero, that is $B_M=0$. Denoting the nonzero coefficients of the susceptibility tensor as

$$-\chi(\pm\,\pm)=a_\pm, \quad \chi_a(\pm\,\pm\,\mp\,\pm)=b_\pm, \quad \chi_a(\pm\,\pm\,\pm\,\mp)+\chi_a(\pm\,\mp\,\pm\,\pm)=c_\pm. \quad (53)$$

The nonlinear equations of interest are

$$2ik^{-1}\frac{\partial A_\pm}{\partial z}+2i\omega^{-1}\frac{\partial A_\pm}{\partial t}=A_\pm\{a_\pm-b_\pm|A_\pm|^2-c_\pm|A_\mp|^2\}. \quad (54)$$

These equations indicate that a wave of given polarization must be incident to be amplified, that is right circular does not generate left circular.

Circularly Polarized Incident Radiation

If the incident traveling wave is right circular and only steady-state operation is of interest, Eq. (54) reduces to

$$2i(dA_+/dz)=kA_+(a_+-b_+|A_+|^2). \quad (55)$$

Making the substitution

$$A_+=Ue^{iu},$$

and writing a_+ and b_+ in the form

$$a_+=a_+'+ia_+'', \quad (56)$$

the real and imaginary parts of Eq. (55) yield an equation for the amplitude U and an equation for the phase u. Direct integration yields

$$U^2(z)=U_0{}^2e^{\alpha z}\times[1+U_0{}^2(b_+''/a_+'')(e^{\alpha z}-1)]^{-1}\xrightarrow[z\to\infty]{}(a_+''/b_+''), \quad (57)$$

where α is the linear gain term $\alpha=ka_+''=\omega c^{-1}a_+''$. Direct integration of the equation for du/dz yields an accumulated phase of

$$u=-\tfrac{1}{2}(\omega/c)\int_0^z dz(a_+'-b_+'U^2). \quad (58)$$

Saturation occurs for large z and the amplitude is limited by the ratio of a_+''/b_+''. a_+'' or $\chi''(+\,+)$ is the Doppler gain curve as given by Eq. (33) and has the frequency dependence of a sum of Z functions which differ by the term $(\gamma_a m_a-\gamma_b m_b)\mathfrak{B}$ for each m_a. These appear as displaced Gaussian gain curves. b_+'' or $\chi_a''(+\,+\,-\,+)$ requires detailed evaluation of terms sensitive to ω and magnetic field sensitive equations of the type given by Eq. (36), or (38a) and (38b). For large Doppler broadening b_+'' is approximately constant and the gain curve has the frequency dependence of $a_+''(\omega)$, that is the Doppler line. This is apparent from Eq. (38a) which gives the form of b_+'' or $\chi(+\,+\,-\,+)$ of line 9 of Table I. Since the M's are all equal, $m_a-m_a'=0$ and $m_b-m_b'=0$ and b_+'' is not sensitive to the input fre-

quency or magnetic fields. This is not true when the $\langle G_1\rangle$ and $\langle G_3\rangle$ yield non-negligible coefficients for the other two integrals in the intermediate region of Doppler broadening.

The accumulated phase u is zero for $\omega=\omega_{ab}$ and zero magnetic field, and is linear in the field for weak magnetic fields. If the Z functions in Eq. (33) are expanded in terms of the magnetic field $\mathfrak{B}$, at line center the real part of Z is of the form,

$$Z'\simeq -2D^{-1}(1+\pi^{1/2}\Gamma_{ab}/D)(\gamma_a m_a-\gamma_b m_b)\mathfrak{B}. \quad (59)$$

Summation of the C-G coefficients over m_a in Eq. (33) with Z replaced by the approximate value of Z' given in Eq. (59) yields a term proportional to a_+'. b_+' follows from an analysis of the integrals in Eq. (29a) and for extreme Doppler broadening is almost zero. Beam-splitting techniques could be used to measure the phase shift as a function of the magnetic field $\mathfrak{B}$ and yield some information concerning the γ's for the excited atomic levels. This equation differs from the oscillator equation in that it does not depend on the bandwidth of a cavity.

$a_\pm'$ is a measure of the deviation of the index of refraction from unity. If the nonlinear term is included only the accumulated phase u can be defined.

Elliptical Polarization for the Incident Radiation

If the incident radiation has elliptical polarization and frequency ω, then both $A_\pm$ are needed in Eq. (54). Again steady state is assumed and the time-dependent term omitted. For convenience let

$$A_+=Ue^{iu} \quad \text{and} \quad A_-=Ve^{iv},$$

and the amplitude aspects of Eq. (54) are given by

$$dU^2/dz=k[a_+''-b_+''U^2-c_+''V^2]U^2=kQ, \quad (60a)$$

$$dV^2/dz=k[a_-''-b_-''V^2-c_-''U^2]V^2=kP. \quad (60b)$$

These may be examined for stable solutions by a standard procedure of phase trajectories[20] and the phase diagrams are similar to those used by Lamb[1] in discussing the mode coupling in a linear oscillator or by Heer[21] in discussing the coupling between clockwise and counter clockwise traveling waves in the photon rate gyroscope. The phase trajectory equation

$$dV^2/dU^2=P(U,V)/Q(U,V) \quad (61)$$

has singularities at $P=Q=0$. The straight lines for $P=0$ and $Q=0$ are shown in Fig. 3 and the singularities occur at $U^2=0$, $V^2=a_-''/b_-''$; $U^2=a_+''/b_+''$, $V^2=0$; and at the intersection of the two lines. The path of the phase trajectory is horizontal as it crosses the line $P=0$ and vertical as it crosses $Q=0$. The intersection occurs and is stable if

$$b_+''/c_-''>a_+''/a_-''>c_+''/b_-'', \quad (62a)$$

and both U^2 and V^2 can coexist with amplitudes

$$U^2=(a_+''b_-''-a_-''c_+'')/(b_+''b_-''-c_-''c_+'')$$
$$\text{and} \quad (62b)$$
$$V^2=(a_-''b_+''-a_+''c_-'')/(b_+''b_-''-c_-''c_+'').$$

Otherwise one of the other singularities is approached and only the right or the left circularly polarized waves leaves the amplifier under saturation conditions. The ratio of the linear gain term a_+''/a_-'' is unity in the absence of a magnetic field and remains the order of unity in weak magnetic fields. Of course in strong fields the ratio may be made larger or smaller than unity.

If $j_a=1$ and $j_b=0$ or $j_a=1$ and $j_b=1$ then b_+''/c_+'' is unity in zero magnetic field and of the order of unity in weak magnetic fields, and there is a tendency for the intersection to become a saddle point. This is apparent from Table I by observing that $b_\pm$ depends on entry 9 or 8 and $c_\pm$ on entry 7+18 or 6+19. Small changes can make this saddle point stable or unstable. $j_a=2$ and $j_b=1$ or $j_a=3$ and $j_b=2$ are highly stable at the intersection. $j_a=2$ and $j_b=2$ is unstable and implies that such a system in a zero or weak axial magnetic field quenches one of the two states of polarization as saturation is approached. This surprising result would be interesting to examine experimentally.

The accumulated phase difference u or v for right or left circular polarization is given by direct integration of du/dz and dv/dz and the phase u after traveling distance z is

$$u=-\tfrac{1}{2}\omega c^{-1}\int_0^z dz(a_+'-b_+'U^2-c_+'V^2), \quad (63)$$

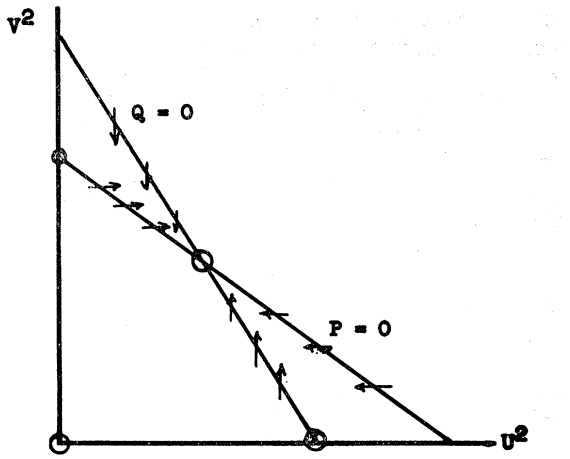

FIG. 3. Phase plane for $dV^2/dU^2=P/Q$ where
$P=[a_1-b_1V^2-c_1U^2]V^2$ and $Q=[a_2-b_2U^2-c_2V^2]U^2$.
The straight lines are for $P=0$ and $Q=0$ and the singularities are indicated by ○. The intersection is stable if $b_2/c_1>a_2/a_1>c_2/b_1$ and is shown above. Figures 3, 4, and 5 of Ref. 1 give detailed phase-plane diagrams for this equation.

[20] A. Andronow and A. Witt, Archiv. fur Elecktrotechnik 24, (1930); N. Minorsky, *Non-Linear Mechanics* (Edwards Brothers, Inc., Ann Arbor, Michigan, 1947), p. 341.

[21] C. V. Heer, in *Proceedings of Colloque Sur Les Gyroscopes Avances, Paris, 1964* (Sci. et Techniques de L'Armement, to be published); and *Proceedings of Symposium on Unconventional Inertia Sensors for 1964* (Republic Aviation Corp., Farmingdale, N. Y., 1965), pp. 221–231.

with a similar expression for v. Reference to Table I indicates that in the absence of hyperfine splitting and for ω near line center the $c_\pm'$ term is most sensitive to the magnetic field. A variation in the magnetic field $\mathfrak{B}$ will cause c' to be strongly dependent on the width Γ_a, Γ_b, and Γ_{ab} as is apparent by examination of Eqs. (38a) and (38b). The polarization of the radiation leaving the amplifier depends on U^2, V^2, u, and v. No simple conclusions can be reached for atoms with j values other than 0 and 1 and in general a detailed analysis is necessary.

The experiment of Hotz[22] on magnetic effects which were interpreted as bandwidth narrowing in a 3.39-μ saturated amplifier provides an interesting example for this theory. The 3.39-μ line of neon is the transition $(5s'[\frac{1}{2}]_1 \rightarrow 4p'[\frac{3}{2}]_2)$. This is a $j_a=1$, $j_b=2$ transition. In zero magnetic field $b_+''/c_-''=b_-''/c_+''=46/22$ and $a_+''/a_-''=1$ and the stability condition of Eq. (62) is met. Right and left circularly polarized waves can coexist in this maser medium with zero or weak axial magnetic field. The wave after traveling a distance z in the amplifying medium is of the form

$$(\hat{e}_- Ue^{iu}+\hat{e}_+ Ve^{iv})\exp i(kz-\omega t)+\text{c.c.} \tag{64}$$

For a saturated wave $U^2\approx V^2$ and if the incident wave is linearly polarized $U^2\approx V^2$ throughout the amplifier.

For large Doppler broadening only the $\langle G_2\rangle$ terms in Eq. (29a) are large and the problem can be examined in some detail. Intermediate Doppler broadening requires the retention of many more terms and is not examined here. Using the M values indicated in Table I for entries 9 or 8 for $b_\pm$ and entries 7+18 or 6+19 for $c_\pm$ with Eq. (38a) yields for $\omega=\omega_{ab}$

$$b_\pm=\text{const}(46)(2\Gamma_{ab}^{-1})(\Gamma_b^{-1}+\Gamma_a^{-1}), \tag{65a}$$

$$\begin{aligned} c_\pm=\text{const}\{&[\mp i2\gamma_a\mathfrak{B}+2\Gamma_{ab}^{-1}]^{-1}\\ &\times[\Gamma_b^{-1}+(\mp i2\gamma_a\mathfrak{B}+\Gamma_a)^{-1}]\\ &+21[\mp i2\gamma_b\mathfrak{B}+2\Gamma_{ab}^{-1}]^{-1}\\ &\times[(\mp i2\gamma_b\mathfrak{B}+\Gamma_b^{-1})^{-1}+\Gamma_a^{-1}]\}; \end{aligned} \tag{65b}$$

$c_\pm$ is primarily dependent on the energy level with $j_b=2$.

These expressions indicate that $b_\pm''$ is independent of the magnetic field and $b_\pm'=0$. In zero field $a_\pm'$ and $c_\pm'$ are zero and in weak fields grow linearly with the field. The amplitude of the wave is approximately given by $U^2\approx V^2\approx a''/(b''+c'')$. As the magnetic field is increased c'' should decrease and the amplitude show an increase. If the power output is observed with an unpolarized detector a field sensitive "dip" should occur at $\mathfrak{B}=0$. For larger fields the curve takes on the magnetic field dependence of the linear gain a''. Examination of a_+' and c_+' indicates that for $U^2\approx V^2$,

$$u=-v,$$

and within the validity of these approximations Eq. (64) may be written for $\hat{e}_y$ polarization of the radiation incident on the amplifier as

$$U[\hat{e}_x \sin u-\hat{e}_y \cos u]\sin(kz-\omega t). \tag{66}$$

The phase u follows from Eq. (63) with $b_+'=0$. If a linear polarizer making angle θ with $\hat{e}_y$ is placed in the path, a detector which is not sensitive to polarization measures a signal proportional to

$$\begin{aligned} \text{Signal}\propto U^2[&\sin^2\theta \sin^2 u+\cos^2\theta \cos^2 u\\ &-2\sin\theta\cos\theta \sin u \cos u]. \end{aligned} \tag{67}$$

This explains the central shape of all four curves observed by Hotz for $\theta=0$, $\pm 45°$, $90°$ as u is varied by changing the axial magnetic field. c_+' approaches a maximum value as $\mathfrak{B}$ is increased and this can be determined in terms of the γ's and Γ's in Eq. (65b). This maximum occurs approximately at $2\gamma_b\mathfrak{B}\approx\Gamma$ if the Γ's are almost equal. The absence of a 'dip' at line center for $\mathfrak{B}=0$ and the narrowing of the line as the magnetic field is increased can be explained if $\langle G_1\rangle$ and $\langle G_3\rangle$ contribute to b_+'', that is intermediate Doppler broadening occurs. The presence of more than one isotope can remove the dip.

9. AMPLIFIER WITH TRANSVERSE MAGNETIC FIELD

If the magnetic field is transverse to the direction of the traveling wave, that is Z is perpendicular to z and along x, the polarization is given by

$$\begin{aligned} \epsilon_0^{-1}P_{\alpha'''}=&S(-M''',\alpha''')S^*(-M''',\alpha)\\ &\times\chi(-M''',-M''')A_\alpha+(-)^{M'}S(-M''',\alpha''')\\ &\times S^*(-M,\alpha)S(-M',\alpha')S^*(-M'',\alpha'')\\ &\times\chi(-M''',-M,M',-M'')A_\alpha(A_{\alpha'})^*A_{\alpha''}, \end{aligned}$$

where $S(0,x)=1$ and $S(\pm,y)=\pm 2^{-1/2}$. The details of this transformation are given in Appendix I. Using abbreviated notation, P_x may be written as

$$\begin{aligned} -\epsilon_0^{-1}P_x=&a_xA_x-b_xA_xA_x^*A_x\\ &-c_xA_xA_y^*A_y+d_xA_yA_x^*A_y, \end{aligned} \tag{68}$$

and P_y follows by permuting the indices x and y. The coefficients are given in terms of $\chi(-M''',-M,M',-M'')$ by

$$\begin{aligned} &a_x=-\chi(0\,0), \quad a_y=-\tfrac{1}{2}\{\chi(+\,+)+\chi(-\,-)\},\\ &b_x=+\chi(0\,0\,0\,0)=+(1),\\ &b_y=-\tfrac{1}{4}\{(7)+(9)+(18)+(6)+(8)+(19)\},\\ &c_x=-\tfrac{1}{2}\{(2)+(4)+(15)+(17)\},\\ &c_y=+\tfrac{1}{2}\{(3)+(14)+(5)+(16)\},\\ &d_x=+\tfrac{1}{2}\{(10)+(13)\},\\ &d_y=-\tfrac{1}{2}\{(12)+(11)\}, \end{aligned} \tag{69}$$

where (7) refers to $\chi(+\,+\,+\,-)$ given by entry No. 7 in Table I, etc. It is convenient to use this table to keep

[22] D. F. Hotz, Appl. Phys. Letters **6**, 130 (1965); Appl. Optics 4, 527 (1965).

in mind the diagrams or integrals giving rise to each term in the electric susceptibility. The effect of magnetic fields and tuning on the coefficient may be visualized in this manner without performing actual calculations. The coefficients are selected such that a_x'', b_x'', $\cdots$, c_y'' are positive constants in zero magnetic field. The sign of d_x'' and d_y'' depends on the j values for the levels and are of opposite sign in zero field.

Equations (47) and (52) may now be used with $\alpha=x$ or y and the equation of motion for A_x is

$$2ik^{-1}\frac{\partial A_x}{\partial z}+2i\omega^{-1}\frac{\partial A_x}{\partial t}=a_xA_x-b_xA_xA_x^*A_x - c_xA_xA_y^*A_y+d_xA_yA_x^*A_y. \quad (70)$$

A similar equation may be obtained for y by permuting x and y. In zero magnetic field $a_x''=a_y''$, $b_x''=b_y''$, etc., and the equations may be placed in the same form as in the previous section. This is expected, since in zero magnetic field the expressions must be independent of the choice of the axis of quantization. In zero magnetic field the coefficients are all imaginary at line center, that is, $a_x'=0$, $b_x'=0$, etc., and only affect the gain. As the magnetic field is increased the real terms a_x', b_x', etc., grow linearly with the magnetic field and contribute to the phase term. Large fields affect both the gain and phase aspects.

The arguments used in the previous section are not as useful as in that section. Substitutions of the type $A_x=U\exp iu$ do not cancel the phase in terms of the type $A_yA_x^*A_y$ and this adds to the complexity of the problem. d_x'' and d_y'' are third-order terms which provide gain rather than saturation for either the A_x or A_y component. This sign depends on the j values and is related to the earlier observation that $j_a=2$, $j_b=2$ is unstable for elliptical polarization. Nonlinear problems of this type will be considered elsewhere. A strong magnetic field along the x direction has less effect on the linear gain term $a_x''=\chi''(0,0)$ than on a_y'' and near the line center the linear gain is larger for the x-polarization. In large fields the gain can be made dominant at one pair of transitions for the linear gain. This may be either x or y depending on the incident frequency. Table I reduces to one of 3 important diagrams, that is 1, 8, or 9 for large magnetic fields. If $A_y=0$ or $A_x=0$ for the wave incident on the amplifier, the analysis is similar to the analysis of Eq. (55) and yields Eqs. similar to (57) and (58).

Amplifier with Two Input Frequencies ω and $\omega+\Delta\omega$

If the amplifier has two input frequencies differing by $\Delta\omega$, then the input wave incident on the amplifier has coefficients of the type

$$A_\pm=A_{1\pm}+A_{2\pm}\exp i(\Delta kz-\Delta\omega t). \quad (71)$$

If the magnetic field is axial Eq. (54) may be used for the analysis of the amplified wave. Both the time and space derivatives are needed for this analysis and this example is introduced to show the general type solutions which the left-hand side of Eq. (54) and the linear gain term can admit. If $\Delta\omega$ is less than the natural width a complex nonlinear problem results.

10. LINEAR OSCILLATORS

Equation (52) may be used for the general linear oscillator with arbitrary magnetic field. Both $E_\alpha(k,\omega)$ or A_α and $E_\alpha(k,-\omega)$ must now be included in $P_\alpha(k,\omega)$ in order to handle traveling waves in the $+z$ direction. The third-order contribution to $P(k,\omega)$ must include the electric susceptibility tensors χ_a, χ_b and χ_c. χ_b and χ_c couple the waves traveling in the $\pm z$ directions. χ_a, χ_b, and χ_c differ through the Doppler effect or $\langle G\rangle$ coefficients in Eq. (29).

In the analysis of the linear oscillator by Lamb[1] terms of the type $\partial A_\alpha/\partial z$ were regarded as negligible and a simple standing wave or normal mode solution introduced. The time-dependent terms gave a measure of the growth of this mode. Losses were introduced by a simple cavity damping term. A standing-wave solution requires

$$\mathbf{E}(k,\omega)=\mathbf{E}^*(k,-\omega) \quad \text{or} \quad \mathbf{A}=\mathbf{B}^*, \quad (72)$$

and reduces the eight equations implied by Eqs. (52a) and (52b) to four equations. This condition will occur for metallic reflecting mirrors and no polarizing devices in the path or in a transverse magnetic field with Brewster-angle windows. The later case may be treated by two equations since $A_x=B_x^*$ and $A_y=B_y=0$ and Lamb has discussed this case in detail. Since $cdt=dz$, there is little difference between this oscillator and the discussion of the linear amplifier in the saturation region. The resulting nonlinear problem is similar to the discussion following Eq. (55).

Linear Oscillator with Axial Magnetic Field

The more general case of an axial magnetic field and perfect reflecting mirrors requires the analysis of a problem similar to the saturated amplification of an elliptically polarized wave. If an approximate normal mode $(2d)^{-1/2}\exp i2\pi qz/2d$ is used for a cavity with mirror spacing of d, the normal mode equations for the qth mode are of the form for the

$$2i(dA_\pm/dt) = \omega_cA_\pm[a_\pm-iQ^{-1}-b_\pm|A_\pm|^2-c_\pm|A_\mp|^2]. \quad (73)$$

Q has been introduced to account for simple cavity losses and ω_c is the cavity frequency. Since standing waves require that

$$A_p=B_p^\dagger, \quad (74)$$

it is apparent that all products in Eq. (29a) are of the form $A_{-p}A_{p'}{}^\dagger A_{-p''}$ and $P_\pm^{(3)}$ depends on only the field terms $A_\pm|A_\pm|^2$ and $A_\pm|A_\mp|^2$. The a, b, c coefficients are given by Eqs. (28a) and (29a). b and c depend on all

coefficients in Eq. (29a) or on χ_a, χ_b, χ_c and the effect of magnetic fields and cavity frequency, $\omega_c=\omega$, is different than in the amplifier. The discussion of the nonlinear problem is the same as used for Eqs. (60a) and (60b) with $dz=cdt$. Equation (61) follows and the stability condition for oscillation of both right and left circularly polarized waves is given by Eq. (62a) and the amplitude of the two waves by Eq. (62b). For large Doppler broadening both Eqs. (38a) and (38b) are needed for b and c. Only Eq. (38a) is required for a linear amplifier. Equation (38b) is sensitive to the cavity frequency ω_c relative to the line center ω_{ab}, and for $\mathfrak{B}=0$ and a single isotope leads to the cavity tuning "dip" suggested by Lamb.[1] The form of the amplitudes are given by Eq. (62b) by replacing a_+'' by $a_+''-Q^{-1}$. In zero magnetic field and near line center this reduces to

$$U^2\approx V^2\approx(a''-Q^{-1})/(b''+c''). \tag{75}$$

The cavity tuning dip is introduced by the presence of both Eqs. (38a) and (38b) in the b'' and c'' coefficients.

In Table I $b_\pm$ depends on entry 8 or 9 and $c_\pm$ on entry 7+18 or entry 6+19. In zero magnetic field the ratio of b_+''/c_+'' is given by $\sum\{\text{C-G}\}$. Combining these values with the stability condition which is given by Eq. (62a), gives the condition under which right and left circular polarization can coexist in zero or weak magnetic fields. $a_+''/a_-''=1$ for zero magnetic field and $\omega_c=\omega_{ab}$. Under these same conditions $b''/c''=1$ for $(j_a=1,\ j_b=0)$ or $(j_a=1,\ j_b=1)$ and a saddle point exists. $b''/c''=46/22$ for $(j_a=2,\ j_b=1)$ and 371/247 for $(j_a=3,\ j_b=2)$, and the intersection in Fig. 3 is quite stable. $b''/c''=26/42$ for $(j_a=2,\ j_b=2)$ and right and left circular polarization cannot coexist in this oscillator in zero field and at line center. This is similar to the conclusion reached for the linear amplifier with an axial magnetic field. The ratios given above remain the same if the j values of j_a and j_b are interchanged.

The discussion remains valid if the hyperfine splitting is large and a single set of F_a and F_b levels can be used. Fowles and Jensen[23] examined the maser oscillation between the hyperfine levels in iodine and found that $(F_a=\frac{7}{2},\ F_b=\frac{9}{2})$ gave the only maser line. If only one polarization is amplified for $(F_a=\frac{7}{2},\ F_b=\frac{7}{2})$, then Brewster-angle windows can introduce a 50% loss and prevent oscillation. It is premature to suggest this effect for their experiment, but it does indicate that this rather surprising dependence of stability on the j or F values must be considered in future experimental arrangements.

The phase is given by an equation similar to Eq. (63), and

$$\begin{aligned}\dot{u}&=-\tfrac{1}{2}\omega_c(a_+'-b_+'U^2-c_+'V^2),\\ \dot{v}&=-\tfrac{1}{2}\omega_c(a_-'-b_-'V^2-c_-'U^2),\end{aligned} \tag{76}$$

and the frequency of oscillation is

$$\omega_+=\omega_c-\dot{u}\quad\text{and}\quad\omega_-=\omega_c-\dot{v}. \tag{77}$$

[23] G. R. Fowles and R. C. Jensen, Phys. Rev. Letters **10**, 347 (1965).

The magnitude of the frequency shift depends on U^2 and V^2 and these are in principle known for an oscillator. In zero magnetic field and at line center $a'=b'=c'=0$ and the frequencies are the same. As discussed earlier, both U and V can occur or they can occur separately. At line center or $\omega_c=\omega_{ab}$ these coefficients increase linearly with the magnetic field and have opposite signs. $a_+'=-a_-'$, $b_+'=-b_-'$, and $c_+'=-c_-'$. The difference in frequency between right and left circular polarization is given by

$$\omega_+-\omega_-=\dot{v}-\dot{u}. \tag{78}$$

Since $\dot{u}$ and $\dot{v}$ have opposite signs a beat frequency proportional to the axial magnetic field will occur and has been observed experimentally.[4,24] In an ideal system a $(j_a=2,\ j_b=1)$ transition should have this frequency difference down to zero magnetic field. Less stable lines will tend to "lock" together due to the lack of ideality in the system. All terms in Eqs. (28a) and (29a) must be included in correlating the beat frequency between the two modes with the γ and Γ values. If the oscillator is not at line center, then the a, b, c coefficients must be modified to take the frequency difference $\omega_c-\omega_{ab}$ into consideration and the previous discussion must be modified. At line center b_+' and c_-' reach a maximum value at a field strength $\gamma\mathfrak{B}\approx\Gamma$, and the quantity $(\dot{u}-\dot{v})$ at high power levels should show a similar maximum.

The linear amplifier showed a dip at line center as the magnetic field was varied. The oscillator tuned for $\omega_c=\omega_{ab}$ has an output which is dependent on $U^2\approx(a''-Q^{-1})/(b''+c'')$. Equation (38b) must now be included in the b'' and c'' and for large Doppler the magnetic field dependence of the amplitude is different from that for the amplifier. Culshaw and Kannelaud[4] have examined this beat frequency in some detail and found a dip at zero field. Brewster-angle windows were used with an axial magnetic field in their experiment and this complicates the analysis of the problem. The observed shape of their power output curve is in agreement with that expected for U^2.

Linear Oscillator with Arbitrary Magnetic Field

The linear oscillator with arbitrary magnetic field and with polarizing devices in the path is described by the basic Eq. (52). It does not appear possible to define a simple normal mode with a simple loss term under these conditions for a cavity. This is apparent by considering a wave in a linear amplifier with an axial field. A linearly polarized wave has it plane rotated during amplification and the loss at the next Brewster angle window depends on the amount of rotation. The problem becomes quite complex and is only formulated here. Fork and Patel[25] have experimentally examined the characteristics of a maser in very strong transverse and axial magnetic fields

[24] R. D. Graft, Ph.D. dissertation, Ohio State University, 1965 (unpublished).

[25] R. L. Fork and C. K. N. Patel, Appl. Phys. Letters **2**, 180 (1963).

and their results can be described in terms of the strong field a and the b and c coefficients which depend on diagrams 1 and 8 or 9 in Table I.

11. CLOSED PATH OSCILLATORS AND THE PHOTON RATE GYROSCOPE

The equations for the electromagnetic fields in a closed path oscillator[7] with a simple transverse polarization following Eq. (45) are

$$2ik^{-1}\frac{\partial A_\alpha}{\partial z}+2i\omega^{-1}\frac{\partial A_\alpha}{\partial t}+2[c^{-1}(\boldsymbol{\Omega}\times\mathbf{r})\cdot\hat{a}_z]A_\alpha = -\epsilon_0^{-1}P_\alpha(k,\omega), \quad (79a)$$

$$2ik^{-1}\frac{\partial B_\alpha}{\partial z}-2i\omega^{-1}\frac{\partial B_\alpha}{\partial t}-2[c^{-1}(\boldsymbol{\Omega}\times\mathbf{r})\cdot\hat{a}_z]B_\alpha = -\epsilon_0^{-1}P_\alpha(k,-\omega). \quad (79b)$$

Both A_α and B_α are needed for the $\pm z$ traveling waves and all eight nonlinear equations are needed for a detailed solution of the problem. The effect of rotation on the cavity has been included by giving space a pseudo-index of refraction in this short-wavelength limit of the form

$$n=n_0+c^{-1}(\boldsymbol{\Omega}\times\mathbf{r})\cdot\hat{a}_z+O(\Omega^2),$$

where Ω is the angular rate of rotation, $\hat{a}_z$ is the direction of the optical path, and $\mathbf{r}$ is the distance from an origin. This is equivalent for $n_0=1$ to a contribution to the polarizability $(n^2-1)\approx 2c^{-1}[(\boldsymbol{\Omega}\times\mathbf{r})\cdot\hat{a}_z]$. Justification for this procedure and modifications for $n_0\neq 1$ are given in Ref. 7.

If the cavity structure permits a simple separation of the time and space variables and a normal mode expansion is made using $L^{-1/2}\exp i2\pi qz/L$ as basis functions, the field equations may be integrated around the closed path of length L to yield the time dependence of the normal mode with index q. The correction for rotation may be written as

$$2c^{-1}\boldsymbol{\Omega}\cdot\int_0^L dz(\mathbf{r}\times\hat{a}_z)/L=4\Omega c^{-1}\times[(\text{enclosed area})/L]\cos\theta=g\Omega.$$

θ is the angle between the normal to the plane formed by the closed optical path and $\boldsymbol{\Omega}$, and g is a geometry factor for the structure. Then the normal mode equations are

$$2i\omega^{-1}(dA_{\alpha q}/dt)+g\Omega A_{\alpha q}=-\epsilon_0^{-1}P_{\alpha q}(k,\omega), \quad (80a)$$

$$-2i\omega^{-1}(dB_{\alpha q}/dt)-g\Omega B_{\alpha q}=-\epsilon_0^{-1}P_{\alpha q}(k,-\omega); \quad (80b)$$

$P_{\alpha q}(k,\omega)$ is the same as $P_\alpha(k,\omega)$, with $k=2\pi q/L$.

These equations have been examined in some detail by Heer[21] for A_x and B_x unequal to zero and $A_y=B_y=0$. Equations (80a) and (80b) reduce to four equations of the form

$$2i(dA_x/dt)=\omega_c A_x[-g\Omega+a-iQ^{-1}-b|A_x|^2-c|B_x|^2], \quad (81a)$$

$$-2i(dB_x/dt)=\omega_c B_x[+g\Omega+a^*+iQ^{-1}-b^*|B_x|^2-c^*|A_x|^2]. \quad (81b)$$

With $A_x=U\exp iu$ and $B_x=V\exp iv$, these reduce to the same form of nonlinear equations as used in the discussion of Eqs. (60a) and (60b) and shown in Fig. 3. The stability equation for the coexistence of clockwise traveling waves is given by Eq. (61) and the amplitudes of these waves by Eqs. (62a) and (62b) with a'' replaced by $(a''-Q^{-1})$. The linear gain coefficient is given by Eq. (28a) and

$$a=-\chi(0,0). \quad (82)$$

From Eq. (A5) in Appendix I, b and c depend on $\chi(0\,0\,0\,0)$. There is no dependence on the m_a, m_b, etc., coefficients and the sum over m_a is just $(2j_a+1)$ times the frequency dependent integral. For atoms at rest $\langle G_1\rangle=\langle G_2\rangle=\langle G_3\rangle$ and

$$c=2b=\chi_a(0\,0\,0\,0). \quad (83)$$

For extreme Doppler motion only the $\langle G_2\rangle$ integrals are appreciable and

$$b\approx\text{const}(\Gamma_{ab})^{-1},\qquad c\approx\text{const}[i(\omega_{ab}-\omega_c)+\Gamma_{ab}]^{-1}. \quad (84)$$

Clockwise and counter-clockwise traveling waves can coexist in this closed path maser if $b''/c''>1$. This occurs for large Doppler broadening if the cavity is tuned away from line center ω_{ab}. Line center is a saddle point. Atoms at rest are unstable at the intersection in Fig. 3. The frequency separation between clockwise and counter-clockwise traveling waves is

$$\omega_{cw}-\omega_{ccw}=\dot{u}+\dot{v}=\omega_c g\Omega, \quad (85)$$

and in this ideal system does not "lock" to a single frequency. The effects of non-ideal conditions are considered in some detail in Ref. (21).

The general case of arbitrary magnetic field and polarizing mirrors or Brewster angle windows requires the use of all eight equations implied by Eqs. (79a) and (79b) and appears to be a rather formidable problem. The discussion of these simpler examples gives some measure of the problems involved.

12. PRESSURE AND OTHER LINE BROADENING

Equations (10) and (11) or (28) and (29) may be corrected for pressure effects in an approximate manner by regarding the energy level spacing ω_{ab} to be perturbed during the collision. The statistical average of the perturbing effect can be taken into account in part by making ω_{ab} complex. Changing Γ_{ab} in Eqs. (28) and to (29)

$$\Gamma_{ab}\rightarrow\Gamma_{ab}+\omega_{ab}'' \quad (86)$$

can be used to introduce a pressure broadening term into the theory.[26,27]

An inhomogeneous magnetic field over the maser length could be used to broaden the line. In this formulation atoms in spatial element $d\mathbf{r}$ would respond to the magnetic field $\mathfrak{B}$ at their position in the tube. The overall gain of the amplifier would depend on an integration over the length of the tube and would incorporate the variation in the gain introduced by the magnetic field variation of $\omega(m_a m_b)-\omega$ over the amplifier length.

13. CONCLUSIONS

The phenomenological integral equation for the density matrix which is given by Eq. (3) has provided a useful model for describing the response of an atom to an external perturbation. An iteration solution of this integral equation is used to obtain the first and third order contributions to the electric polarization vector and these are given by Eqs. (10) and (11). Equations (10) and (11) are sufficiently general that they can be used in the discussion of a multimode problem in which the modes are well separated in frequency or are within the natural width of the line and is more general than the development of Lamb.[1] These equations are valid in arbitrary magnetic field. For almost plane monochromatic traveling waves in the $\pm z$ directions, these equations permit the introduction of a macroscopic electric polarization for a volume element $d\mathbf{r}$ and Eqs. (28) and (29) give the first- and third-order contributions to the electric polarization for arbitrary magnetic field direction. A first- and fourth-order tensor are introduced for the electric susceptibility and forms a convenient notation; but in a given problem it is more convenient to substitute Eqs. (51a) and (51b) directly into Eq. (29). The coefficients of the electric field are regarded as slowly varying functions of position and time and the electric field of the waves and the electric field in the polarization are made self-consistent by direct substitution into Maxwell's equation. This leads to the very general Eqs. (52a) and (52b) which may be used for the discussion of amplifiers, oscillators in which normal modes are defined, and in oscillators in which normal modes are not obvious.

This phenomenological model for the macroscopic polarization of a gas has many features in common with earlier approaches. Hanle[28] was the first to apply quantum theory to problems involving specific polarizations involved in the various Zeeman transitions and Breit[29] gave a thorough discussion of dispersion for atoms with hyperfine structure and in magnetic fields. Coherent excitation and its effects on spontaneous emission were considered by Breit and used to discuss the results obtained in level-crossing experiments.[30] Similar considerations were used in the discussion of double resonance.[31] The coherence in these experiments was between the closely spaced magnetic levels and in the theory developed in this paper correspond to the growth of second-order density-matrix elements $\rho^{(2)}(m_a m_a')$ or $\rho^{(2)}(m_b' m_b)$ .This second-order density matrix gives rise to the third-order contribution to the atomic polarization and modifies the stimulated absorption or emission and spontaneous emission. Only the aspects related to stimulated emission were discussed in this paper.

Since all of the problems of interest were in the nonlinear region only a limited number of simple examples could be considered in detail. In these simple examples and for ideal experimental arrangements, the relationship between the experimental observations and the atomic properties were discussed. These examples were not pursued to their full extent, since the correlation of a nonlinear theory with an experiment is better done in a paper in which these aspects are known before the experiment and are subject to study during the experiment. Even so, reasonable agreement was obtained on the magnetic effects observed in the experiment of Hotz[22] with the linear amplifier, Culshaw and Kannelaud[4] and other observers for the linear oscillator,[24,32] and other aspects of linear oscillators.[1,11]

A surprising aspect of this study was the instability of $j_a=j_b$ or $F_a=F_b$ maser amplifiers or oscillators to the two states of circular polarization. Thus a linear amplifier with $j_a=j_b=2$ prefers to amplify either right circular or left circular polarization and will quench one or the other of the two states of polarization. No experimental check is available for this conclusion.

The most complex problem to consider is the closed-path maser which has clockwise and counter-clockwise modes and two states of polarization for each of these modes. The formulation of this paper is adequate to treat this problem and eight nonlinear equations result. This was studied for an ideal system with linear polarization and it was shown that in such a photon rate gyroscope the clockwise and counter-clockwise traveling waves do not "lock" for large Doppler broadening and the beat frequency between the two waves is proportional to the angular rate of rotation Ω.

Although the formulation of this paper includes hyperfine effects by making the modifications indicated in Sec. 6, it again appeared that the discussion of such examples would be better treated in detail with the appropriate experiments. For large hyperfine splitting most of the previous discussions are applicable by

[26] A. Szoke and A. Javan, Phys. Rev. Letters **10**, 521 (1963).
[27] H. Margenau and M. Lewis, Rev. Mod. Physics **31**, 569 (1959).
[28] W. Hanle, Z. Physik **30**, 93 (1924).
[29] G. Breit, Rev. Mod. Phys. **5**, 91 (1933).
[30] F. D. Colegrove, P. A. Franken, R. R. Lewis, and R. H. Sands, Phys. Rev. Letters **3**, 420 (1959).
[31] G. Brossel and F. Bitter, Phys. Rev. **86**, 308 (1952); J. N. Dodd, W. N. Fox, G. W. Series, and M. J. Taylor, Proc. Phys. Soc. (London) **74**, 789 (1959); J. P. Barratt, Proc. Roy. Soc. (London) **A263**, 371 (1961).
[32] I. Tobias, M. Skolnick, R. Wallace, and T. Polanyi, Appl. Phys. Letters **6**, 198 (1965).

changing j to F. For small hyperfine splitting the trace must be modified as indicated in Eq. (42) and all a, b, c coefficients must include the effect of these additional sums.

Very little consideration was given to the effect of the variation of the electric vector or of states of polarization across the maser beam. Since the general equations were developed for a spatial element $d\mathbf{r}$, these effects can in principle be taken into account as the electric polarization or susceptibility is integrated over the spatial normal mode of the cavity.

ACKNOWLEDGMENT

The authors wish to express their appreciation to R. A. Settles for assisting in the preparation of Table I and other related material.

APPENDIX I

The transformation between two complex vectors expressed in terms of the spherical basis set are given by the $D_{Mp}^{(1)}$ or $D(M,p;\alpha\beta\gamma)$ coefficients and the transformation is of the form

$$A_M = D^*(M,p)A_p. \tag{A1}$$

Repeated indices are summed throughout. These coefficients are elements of a unitary matrix and obey the usual rules for a unitary matrix,

$$\begin{aligned} D^*(M,p)D(M'p) &= \delta(M,M'), \\ D^*(M,p)D(M,p') &= \delta(p,p'). \end{aligned} \tag{A2}$$

The elements of $D(Mp)$ are[15]

$$\begin{aligned} D(++) &= D^*(--) = \tfrac{1}{2}(1+\cos\beta)\exp i(\alpha+\gamma); \\ D(+-) &= D^*(-+) = \tfrac{1}{2}(1-\cos\beta)\exp i(\alpha-\gamma); \\ D(0+) &= -D^*(0-) = 2^{-1/2}\sin\beta\exp i\gamma; \\ -D(+0) &= D^*(-0) = 2^{-1/2}\sin\beta\exp i\alpha \end{aligned}$$

and

$$D(00) = \cos\beta.$$

A more general transformation from any two canonical states of polarization α to the M scheme is needed. This is readily accomplished by transferring the states $\alpha = 1, 2, 3$ to the spherical basis $p = 0, \pm 1$ and then to the M basis. Once done, it is most convenient to use a single unitary matrix for the transformation. Repeating Eq. (50), the transformation is

$$A_M = D^*(Mp)A_p = D(Mp)U^*(p\alpha)A_\alpha = S^*(M\alpha)A_\alpha. \tag{A3}$$

D, U, and S are unitary matrices and S gives the direct transformation. Since the wave is transverse the component of A_α with $\alpha = 3$ or $p = 0$ is taken as zero. If $\alpha = 3$ is z, and Z is along x, the spherical bases vectors are related by $D(M,p;90°,90°,180°)$. $U(p,\alpha)$ relates p and x, y. The elements of the combined operator is quite simple and $S(\pm,x) = 0$, $S(0,x) = 1$, $S(\pm,y) = \mp 2^{-1/2}$, $S(0,y) = 0$, and $S(M,z)$ is not used.

Following the transformation given by Eq. (A3) and noting that $A_M{}^\dagger$ transforms as

$$\begin{aligned} A_M{}^\dagger = (-)^M(A_{-M})^* &= (-)^M[S^*(-M,\alpha')A_{\alpha'}]^* \\ &= (-)^M S(-M,\alpha')(A_{\alpha'})^*, \end{aligned} \tag{A4}$$

the general electric polarization vector may be written in terms of the canonical states of polarization α as

$$\begin{aligned} \epsilon_0{}^{-1}P_{\alpha'''}(k,\omega) &= S(-M'''\alpha''')S^*(-M'''\alpha)\chi(-M''', -M''')A_\alpha \\ &+(-)^{M'}S(-M'''\alpha''')S^*(-M\alpha)S(-M'\alpha')S^*(-M''\alpha'')\chi_a(-M''', -M, M', -M''; k,\omega)A_\alpha(A_{\alpha'})^*A_{\alpha''} \\ &+(-)^{M''}S(-M'''\alpha''')S^*(-M\alpha)S^*(M'\alpha')S(M''\alpha'')\chi_b(-M''', -M, M', -M''; k,\omega)A_\alpha B_{\alpha'}(B_{\alpha''})^* \\ &+(-)^{M}S(-M'''\alpha''')S(M\alpha)S^*(M'\alpha')S^*(-M''\alpha'')\chi_c(-M''', -M, M', -M''; k,\omega)(B_\alpha)^*B_{\alpha'}A_{\alpha''}. \end{aligned} \tag{A5}$$

This is written out in detail to emphasize the differences in the transformations for the tensors χ_a, χ_b, and χ_c. The direct transformation of P_M to P_α using Eqs. (51a), (51b), and (29) seems more convenient.

APPENDIX II

If the correction to the Hamiltonian for the hyperfine splitting and magnetic field is written as

$$H_I = h\mathfrak{A}\mathbf{J}\cdot\mathbf{I} - M\cdot\mathfrak{B}, \tag{A6}$$

then the energy-level splitting in magnetic fields which are weak relative to the hyperfine splitting is given by

$$\begin{aligned} &\omega(F_a m_{F_a}, F_b m_{F_b}) - \omega_{ab} \\ &\approx \tfrac{1}{2}\mathfrak{A}_a[F_a(F_a+1) - j_a(j_a+1) - I(I+1)] \\ &\quad -\tfrac{1}{2}\mathfrak{A}_b[F_b(F_b+1) - j_b(j_b+1) - I(I+1)] \\ &\quad +(\gamma_{F_a}m_{F_a} - \gamma_{F_b}m_{F_b})\mathfrak{B}. \end{aligned} \tag{A7}$$

The γ_F's are defined by the reduced matrix elements

$$h\gamma_{F_a} = -(2F_a+1)^{-1/2}(j_a I F_a \| \mathfrak{M}^{(1)} \| j_a I F_a). \tag{A8}$$

$\mathfrak{M}$ is a first-order tensor operator and in weak fields is proportional to the total angular momentum $\mathbf{F}$ and the constant of proportionality is given by the γ's.

In strong magnetic fields corresponding to the Paschen-Back effect the quantum numbers $(m_a m_I)$ replace $(F_a m_{F_a})$. Optical transitions occur between levels with the same m_I values and

$$\begin{aligned} \omega(m_a m_b) - \omega_{ab} &= (\gamma_a m_a - \gamma_b m_b)\mathfrak{B} \\ &\quad +(\mathfrak{A}_a m_a - \mathfrak{A}_b m_b)m_I \end{aligned} \tag{A9}$$

gives a typical term. Equation (40) follows by omitting the hyperfine term which is regarded as small compared to the term depending on the magnetic field. γ_a and γ_b are defined by the reduced matrix given in Eq. (39).

It is apparent from the equations for the reduced matrix elements that

$$\hbar\gamma = g\mu_B, \tag{A10}$$

where μ_B is the Bohr magneton and g is a constant usually between $\frac{1}{2}$ and 2.

Note added in proof. The conditions which were placed on Γ and λ in the integral formulation which is given by Eq. (3) are more restrictive than necessary. C. P. Yang and C. V. Heer have shown that an integral solution of Eq. (2) exists if a part of Γ commutes with H_0. Then Eq. (2) has the integral solution

$$\rho(t) = \int_0^\infty ds\, T^+(s) \times\{\lambda+[(V/i\hbar-\tfrac{1}{2}\Gamma_1)\rho-\rho(V/i\hbar+\tfrac{1}{2}\Gamma_1)]\}_{(t-s)} T(s),$$

where $T(s)=\exp[-(\frac{1}{2}\Gamma_0-i\hbar^{-1}H_0)s]$. No restrictions are placed on λ and Γ_1, and $\Gamma_0>0$ is sufficient.

PHYSICAL REVIEW VOLUME 140, NUMBER 4A 15 NOVEMBER 1965

$2^{1,3}P$, $3^{1,3}P$, and $4^{1,3}P$ States of He and the 2^1P State of Li^+

B. SCHIFF, H. LIFSON, C. L. PEKERIS, AND P. RABINOWITZ
Department of Applied Mathematics, Weizmann Institute of Science, Rehovoth, Israel
(Received 9 June 1965)

A method is outlined for calculating nonrelativistic eigenvalues and wave functions for a two-electron P state of odd parity, and for evaluating the mass polarization and all of the relativistic corrections, apart from the radiative terms. Calculations have been made for the low-lying P states of He and the 2^1P state of Li^+ using up to 560 terms in the expansion of the wave function. The nonrelativistic eigenvalues converge to within an accuracy of from 10^{-4} cm^{-1} to 10^{-2} cm^{-1}. The values of the mass polarization and of the relativistic corrections converge more rapidly than this, so that the total theoretical ionization energy is estimated to be correct to within an error of not more than 10^{-2} cm^{-1}, i.e., considerably less than the experimental error, in the case of all of the states considered. The difference between the theoretical and experimental term values is in no case greater than 0.1 cm^{-1} in absolute magnitude, and is presumed to be due to the contributions from the radiative terms, which have not been included in the calculation. The term value obtained for the 2^1P state of Li^+ confirms the identification of the line at 9581.42 Å in the Li^+ spectrum as belonging to the 2^1S–2^1P transition.

I. INTRODUCTION

IN this paper, we outline a method for determining two-electron atom P-state term values in which the contributions from the mass-polarization and all of the relativistic corrections, apart from the Lamb shift, have been taken into account. Computations have been carried out for the low-lying P states of helium, and the 2^1P state of Li^+, and in each case the results converge to an accuracy exceeding that of the experimental measurements. The same methods have also been used to compute the fine-structure splitting of the 2^3P and 3^3P levels of helium, the results for which have been published previously.[1]

The classical papers of Breit on the angular dependence of a two-electron P-state wave function[2] and the fine-structure splitting of the helium 2^3P level[3] appeared soon after the basic work of Hylleraas on the ground state.[4,5] Subsequent calculations for the P state were mainly performed with the object of determining the fine structure of the ^3P levels, and in contradistinction to the case of the S state, no complete calculation of the relativistic corrections for a two-electron P state has up till now been made, no doubt in view of their greater complexity. We shall therefore give considerable detail in the following exposition.

At the time when the work to be described below was started, the most accurate calculations for a two-electron P state were those of Araki *et al.*[6] and of Traub and Foley.[7] The latter authors were able to obtain a theoretical ionization energy within 15 cm^{-1} of the experimental value for the helium 2^3P state by optimizing the values adopted for the screening constants for the two electrons, and by including 18 terms in the expansion of the wave function. We set out with the aim of computing ionization energies for two-electron P states to an accuracy which would at least match that of the most recent experimental data[8,9] (± 0.03 cm^{-1}). The method used to determine the nonrelativistic eigenvalues and wave functions is an extension of that developed by one of us for excited S states,[10] the wave function being

[1] B. Schiff, C. L. Pekeris, and H. Lifson, Phys. Rev. **137**, A1672 (1965).
[2] G. Breit, Phys. Rev. **35**, 569 (1930).
[3] G. Breit, Phys. Rev. **36**, 383 (1930).
[4] E. A. Hylleraas, Z. Physik **48**, 469 (1928).
[5] E. A. Hylleraas, Z. Physik **54**, 347 (1929).
[6] G. Araki, K. Mano, and M. Ohta, Progr. Theoret. Phys. (Kyoto) **22**, 469 (1959).
[7] J. Traub and H. M. Foley, Phys. Rev. **116**, 914 (1959).
[8] G. Herzberg, Proc. Roy. Soc. (London) **A248**, 309 (1958).
[9] G. Herzberg and H. R. Moore, Can. J. Phys. **37**, 1293 (1959).
[10] C. L. Pekeris, Phys. Rev. **127**, 509 (1962).