Nonlinear Effects in the Resonant Absorption of Several Oscillating Fields by a Gas

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The lowest order nonlinear effects due to saturation in the simultaneous resonant absorption of several oscillating fields by a low-pressure gas are considered. The analysis is based on a perturbation solution of the Boltzmann equation for the density matrix of the gas in a quantum statistical description. Within certain ranges of the parameters involved, it is shown that: (1) When the gas is irradiated by two oscillating linearly polarized fields of different frequencies well within the Doppler-broadened width of the same absorption line, maximum absorption of both fields occurs when the two linear polarizations are perpendicular to each other. (2) When the gas is in a static magnetic field and irradiated by two right-circularly polarized fields at ω_0 and $\omega_0 + \Delta\omega$, and two left-circularly polarized fields at ω_0 and $\omega_0 - \Delta\omega$, all well within the Doppler width of the respective Zeeman components ($\Delta m = +1$ and -1) of the same absorption line, there exists a definite phase relationship among the fields whereby absorption of all four fields is simultaneously at a maximum. Although these effects are quite small in normal spectroscopic work, manifestation of both effects is readily observable in gaseous He-Ne optical masers.

I. INTRODUCTION

IN this paper, we consider the nonlinear effects due to saturation in the simultaneous resonant absorption, or stimulated emission, of several coherent monochromatic radiation fields by a low-pressure gas. This investigation was motivated by the following questions arising in our study on a gaseous He-Ne optical maser.[1,2] In such a maser, the Doppler width of the Ne emission line of interest is considerably broader than the separation between successive resonant frequencies of the radiation cavity. When the maser excitation is strong and the population inversion between the two excited Ne states of interest is much higher than the threshold value required for maser action, coherent oscillation can take place in several longitudinal modes of the cavity. According to the usual linear theory of masers,[3-5] the various oscillations are expected to be independent of each other. Experimentally, however, certain relationships among the various oscillating modes of the maser are observed: (1) In the absence of an external magnetic field, the maser often oscillates in several linearly polarized modes. It is observed[6] that the polarizations in successive modes are at right angles to each other. (2) In another set of observations,[7] in the presence of an external magnetic field applied along the axis of the maser, it is found that the maser could oscillate simultaneously in at least three modes. There exist certain phase relationships among the various oscillations such that no net beat note at the difference frequency

between two successive modes could be detected. (See Sec. V below for more details.) It is the purpose of the present paper to examine the role of the nonlinearity due to the saturation effect in determining the relationships among the various oscillating modes in a gaseous maser. For simplicity, the more compact density-matrix formulation is used in the calculations here.

In brief, when the gas is irradiated by several coherent oscillating fields, the total rate of transition is modulated at the difference frequencies; therefore, the populations of the states of interest are also modulated. If the nonlinear saturation effect is important in the sense that the rate of induced transitions is not negligible compared with the relaxation rate, the absorption processes at the different frequencies of the applied radiation will, therefore, not be independent of each other. The basic problem is to determine the lowest order nonlinear term in the electric dipole moment of a gas induced by the radiation fields. We obtain this from a perturbation solution of the Boltzmann equation for the density matrix of the gas. When the nonlinear effects as discussed above are taken into account, it can be shown that both the absorptive part and the dispersive part of the induced dipole moment of the gas at any frequency would depend upon not only the magnitudes but also the relative orientations of the polarizations or the phase relationships among the fields. In the presence of a population inversion, or a negative temperature, the above effects imply that the negative absorption by the gas, or the gain of the gaseous medium, varies with such relationships among the radiation fields. The experimentally observed polarization or phase relationships would then correspond to the choice where such a negative absorption is the highest; or, alternatively, the threshold for coherent maser action is the lowest.

The use of two radiation fields in masers has been considered in detail in the past by Javan[8] and others.[9-11]

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In most of these considerations, however, one of the radiation fields is of saturation strength and is primarily employed to set up, loosely speaking, a population inversion between two other states of the atomic or molecular systems where maser action is to take place in the form of either a single amplifying or a single oscillating radiation field. Extending the work of Javan,[8] the Doppler effect in a gaseous maser has been considered by Yajima.[12]

Mathematically, the nonlinear effects considered here are not very different from those encountered in the usual optical double-resonance experiments,[13] and in multiple quantum transitions in nuclear magnetic resonance work. In the former case, the systems interacting with the radiation fields are practically isolated. In the latter case, multiple quantum transitions take place in nonisolated spin systems and a variety of relaxation processes must be considered.[14] In the present work, the relaxation process in the gaseous system envisaged here is considerably simpler—only hard collisions dominate. Such a process can be characterized by a single mean collision time in the Boltzmann equation for the density matrix. Furthermore, the effects we consider here involve successive single-photon processes rather than true multiple-photon processes.

Karplus and Schwinger[15] give a general theory of saturation in the absorption of radiation by a gas. There, the applied radiation is a single monochromatic wave. The gaseous system considered in the present work is similar to that considered by these authors; the problem is, therefore, formulated as an adaptation of their general formulation to the situation where the applied radiation consists of several monochromatic components. We restrict our considerations, however, to the case where the radiation field is relatively weak, so that the rate of induced transition is small, but not negligible, compared with the collision rate, in order to facilitate solution of the Boltzmann equation by perturbation techniques. The procedure used is described in the following section.

Two specific examples that need be considered in order to explain the polarization and phase relationships in maser oscillations are given in the succeeding sections. In all these cases, the perturbation calculation is carried out up to the first nonlinear term in the induced dipole moment of the gas. The application of these calculated results to the maser system is discussed in detail in Sec. V.

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II. THE BOLTZMANN EQUATION

The problem here is to determine the resonant part of the average electric dipole moment per molecule, $\mathbf{p}(t)$, of a gas, in which τ is the average time between collisions of a molecule, induced by an applied electric field, $\mathbf{F}(t)$, consisting of several monochromatic components:

$$\mathbf{F}(t) = \sum_l \mathbf{F}^{(l)} \cos(\omega_l t + \theta_l). \tag{2.1}$$

Once this is known, the resonant absorption of the radiation by the gas can be determined readily. The Doppler effect can be taken into account simply by averaging over the translational velocity of the molecules. The Hamiltonian of the gas molecule, including the energy in the external field, is

$$H = H_0 - \sum_l \mathbf{p} \cdot \mathbf{F}^{(l)} \cos(\omega_l t + \theta_l)$$
$$\equiv H_0 + \sum_l V^{(l)} \cos(\omega_l t + \theta_l), \tag{2.2}$$

where H_0 is the Hamiltonian of the isolated molecule and $\mathbf{p}$ is the dipole moment operator. The resonant part of the induced dipole moment is given by

$$\mathbf{p}(t) = \mathrm{Tr}[\mathbf{p}D(t)], \tag{2.3}$$

where $D(t)$ is the resonant part of the density matrix. Using a representation in which H_0 is diagonal and under the usual assumptions,[15] $D(t)$ is determined from the Boltzmann equation

$$\left(\frac{\partial}{\partial t} + i\omega_{mn} + \frac{1}{\tau}\right)D_{mn}(t)$$

$$= -\frac{\rho_m^{(0)} - \rho_n^{(0)}}{\hbar\omega_{mn}} \sum_l \frac{d}{dt} V^{(l)} \cos(\omega_l t + \theta_l)$$

$$-\frac{i}{\hbar} \sum_{kl} [V_{mk}^{(l)} D_{kn}(t) - D_{mk}(t) V_{kn}^{(l)}]$$
$$\times \cos(\omega_l t + \theta_l), \tag{2.4}$$

where

$$\rho^{(0)} = \exp\left(-\frac{H_0}{kT}\right) \bigg/ \mathrm{Tr}\left[\exp\left(-\frac{H_0}{kT}\right)\right] \tag{2.5}$$

is the equilibrium distribution function at the temperature T in the absence of the applied field. The complete derivation of Eq. (2.4) is given in reference 15, with the only difference in the perturbation term V. Here V has several components, $V^{(l)} \cos(\omega_l t + \theta_l)$. This complicates the solution of the equation and it is not feasible to use the same method[10,15] of solution. For the present purpose, it is sufficient to consider the case of moderately weak fields in the sense that the rate of the induced transitions is small, though not negligible, compared with the collision rate, or

$$|V_{mn}^{(l)}|/\hbar\tau^{-1} \ll 1. \tag{2.6}$$

With this restriction, the Boltzmann equation (2.4) can be conveniently solved by standard perturbation techniques. Specifically, we have

$$D_{mn} = D_{mn}^{(1)} + D_{mn}^{(2)} + D_{mn}^{(3)} + \cdots. \tag{2.7}$$

The lowest order term in D_{mn} is linear in $V_{mn}^{(l)}$.

$$D_{mn}^{(1)} = 0 \quad \text{for} \quad V_{mn}^{(l)} = 0 \qquad (2.7\text{a})$$

$$= \Delta\rho_{mn}^{(0)} \int^t \frac{d}{dt'} \Big[\sum_l V_{mn}^{(l)} \cos(\omega_l t' + \theta_l) \Big]$$

$$\times \exp\Big(i\omega_{mn} + \frac{1}{\tau} \Big)(t'-t)dt'$$

$$\text{for} \quad V_{mn}^{(l)} \neq 0, \quad (2.7\text{b})$$

where

$$\Delta\rho_{mn}^{(0)} = -(\rho_m^{(0)} - \rho_n^{(0)})/\hbar\omega_{mn}. \qquad (2.8)$$

Equation (2.7a) includes, obviously, all the diagonal terms of $D^{(1)}$, since in the present representation, V has no diagonal terms. For the subsequent terms, i.e., $j \geq 2$:

$$D_{mn}^{(j)} = -\frac{i}{\hbar} \int^t \sum_{kl} \big[V_{mk}^{(l)} D_{kn}^{(j-1)}(t')$$

$$- D_{mk}^{(j-1)}(t') V_{kn}^{(l)} \big] \cos(\omega_l t' + \theta_l)$$

$$\times \exp\Big[\Big(i\omega_{mn} + \frac{1}{\tau} \Big)(t'-t) \Big] dt'. \quad (2.9)$$

Equations (2.7)–(2.9) constitute a formal series solution of Eq. (2.4); the successive terms in the series are of the order $|V_{mn}^{(l)}|/\hbar\tau^{-1}$ smaller. An additional simplification can usually be made in the application of Eqs. (2.7)–(2.9): Only near resonance terms need be kept in the multiple integrations implied in (2.9).

For the case of a monochromatic radiation field, it can be verified that the procedure outlined here also leads to the well-known expressions[15] for the susceptibility of a gas, with or without saturation. In this case, the perturbation routine is much more cumbersome than the usual method.[10,15] However, in the case where the applied radiation consists of several components at different frequencies, it serves as a simple systematic procedure of obtaining the first few terms in a perturbation expansion of D_{mn}, which is sufficient in view of (2.6).

III. EXCITATION BY TWO LINEARLY POLARIZED FIELDS

Consider a gas of molecular density N, with a nonequidistant discrete spectrum irradiated by two linearly polarized fields of angular frequencies ω_0 and $\omega_0 + \Delta\omega$, where $\Delta\omega \ll \omega_0$ and ω_0 are close to the transition frequency, ω_{12}, corresponding to a particular pair of energy levels, E_1 and E_2 [$E_1 > E_2$, (Fig. 1)], of the gaseous system. The perturbation term in the Hamiltonian, Eq. (2.2), has the form

$$V = -\mathbf{p} \cdot \mathbf{F}^{(a)} \cos\omega_0 t - \mathbf{p} \cdot \mathbf{F}^{(b)} \cos(\omega_0 + \Delta\omega)t$$

$$= V^{(a)} \cos\omega_0 t + V^{(b)} \cos(\omega_0 + \Delta\omega)t. \qquad (3.1)$$

With the form of the perturbation specified, the induced dipole moment of the gas can be determined from the density matrix to be computed through a straight-
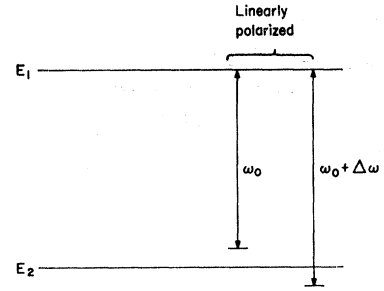

FIG. 1. A two-level system irradiated by two linearly polarized fields.

forward application of Eqs. (2.7)–(2.9). As noted previously, only near resonance terms need be kept, assuming that no other transition frequencies, $\omega_{m'n'}$, are close to ω_0 in the sense

$$|\omega_{12} - \omega_0| \ll |\omega_{m'n'} - \omega_0|. \qquad (3.2)$$

Substituting Eq. (3.1) into Eq. (2.7b), and in view of Eq. (3.2), we obtain for the first approximation

$$D_{12}^{(1)} = -\frac{i\Delta\rho_{12}^{(0)}\tau}{2} \Big[\frac{\omega_0 V_{12}^{(a)} e^{-i\omega_0 t}}{1 + i\Omega_{12}\tau}$$

$$+ \frac{(\omega_0 + \Delta\omega) V_{12}^{(b)} e^{-i(\omega_0 + \Delta\omega)t}}{1 + i\Omega_{12}^{(+)}\tau} \Big], \quad (3.3)$$

where

$$\Omega_{12} = \omega_{12} - \omega_0 > 0,$$

$$\Omega_{12}^{(+)} = \omega_{12} - (\omega_0 + \Delta\omega) < 0; \qquad (3.4)$$

in addition,

$$D_{21}^{(1)} = D_{12}^{(1)*},$$

$$D_{11}^{(1)} = D_{22}^{(1)} = 0. \qquad (3.5)$$

$D^{(1)}$ is linear in $V^{(a)}$ and $V^{(b)}$. The corresponding average dipole moment per molecule of the gas in this approximation, $\mathbf{p}^{(1)}(t)$, is computed from $D^{(1)}$ using Eq. (2.3) and then averaging over the orientation of the molecule, in view of the spherical symmetry of an isolated molecule. The power absorbed from each frequency component of the radiation is proportional to the time average of the scalar product of the radiation field and the rate of change of the corresponding Fourier component of $\mathbf{p}^{(1)}(t)$. This leads to the usual absorption coefficient of the gas, and the absorption processes at the two frequencies of the applied radiation are completely independent of each other; needless to say, they do not depend upon the relative orientation of the polarizations. Such a dependence will come in through the nonlinear terms associated with the higher order terms to be determined using formula (2.9).

For the next order, with Eqs. (3.3)–(3.5), we obtain all the elements of $D^{(2)}$ using (2.9). $D^{(2)}$ is, however, diagonal; it does not contribute directly to $\mathbf{p}(t)$, or $\mathbf{p}^{(2)}(t) = 0$. To obtain the first nonlinear term in $\mathbf{p}(t)$, we must continue on to the next order of approximation,

$D^{(3)}(t)$:

$$D_{12}^{(3)} = V_{12}^{(a)}e^{-i\omega_0 t}\big[\,|V_{12}^{(a)}|^2\alpha_{12}(\omega_0)+|V_{12}^{(b)}|^2\beta_{12}(\omega_0)\big]$$

$$+V_{12}^{(b)}e^{-i(\omega_0+\Delta\omega)t}\big[\,|V_{12}^{(a)}|^2a_{12}(\omega_0+\Delta\omega)$$

$$+|V_{12}^{(b)}|^2\beta_{12}(\omega_0+\Delta\omega)\big]=D_{21}^{(3)*}, \quad (3.6)$$

where

$$\alpha_{12}(\omega_0)=+i\frac{\Delta\rho_{12}^{(0)}\tau^3}{4\hbar^2(1+i\Omega_{12}\tau)}\frac{2\omega_0}{1+\Omega_{12}^2\tau^2}, \quad (3.7a)$$

$$\beta_{12}(\omega_0)=+i\frac{\Delta\rho_{12}^{(0)}\tau^3}{4\hbar^2(1+i\Omega_{12}\tau)}$$

$$\times\Bigg[\frac{2(\omega_0+\Delta\omega)}{1+\Omega_{12}^{(+)2}\tau^2}+\frac{\omega_0+\Delta\omega}{(1-i\Omega_{12}^{(+)}\tau)(1+i\Delta\omega\tau)}$$

$$+\frac{\omega_0}{(1+i\Delta\omega\tau)(1+i\Omega_{12}\tau)}\Bigg], \quad (3.7b)$$

$$a_{12}(\omega_0+\Delta\omega)=+i\frac{\Delta\rho_{12}^{(0)}\tau^3}{4\hbar^2(1+i\Omega_{12}^{(+)}\tau)}$$

$$\times\Bigg[\frac{2\omega_0}{1+\Omega_{12}^2\tau^2}+\frac{\omega_0+\Delta\omega}{(1-i\Delta\omega\tau)(1+i\Omega_{12}^{(+)}\tau)}$$

$$+\frac{\omega_0}{(1-i\Delta\omega\tau)(1-i\Omega_{12}\tau)}\Bigg], \quad (3.7c)$$

$$\beta_{12}(\omega_0+\Delta\omega)=+i\frac{\Delta\rho_{12}^{(0)}\tau^3}{4\hbar^2(1+i\Omega_{12}^{(+)}\tau)}\frac{2(\omega_0+\Delta\omega)}{1+\Omega_{12}^{(+)2}\tau^2}. \quad (3.7d)$$

Again, $\mathbf{p}^{(3)}(t)$ can be determined completely through Eqs. (3.6), (3.7), and (2.3) and averaging over the orientation of the molecule. The explicit dependence of $\mathbf{p}^{(3)}(t)$ on the angle between $\mathbf{F}^{(a)}$ and $\mathbf{F}^{(b)}$, ϕ, comes in through the terms involving the crossed products $V_{12}^{(a)}|V_{12}^{(b)}|^2$ and $V_{12}^{(b)}|V_{12}^{(a)}|^2$. Physically, these crossed products come from the fact that, when the applied coherent radiation consists of two components at different frequencies, the total transition rate is modulated at the difference frequency. The amount of modulation depends upon the relative orientation of polarization of the two fields. If the collision rate is not sufficiently fast as compared with the transition rate, transitions are, therefore, induced between states whose populations are modulated.

Of particular interest in $\mathbf{p}^{(3)}(t)$ are: the component of $\mathbf{p}^{(3)}(t)$ varying harmonically at the angular frequency ω_0 that lies in the direction of $\mathbf{F}^{(a)}$ and the component varying at $\omega_0+\Delta\omega$ that lies in the direction of $\mathbf{F}^{(b)}$, for these alone contribute to the absorption of the radiation fields. It suffices to give one of them here.

$$\mathbf{p}_a^{(3)}(\omega_0)\equiv\big[\mathbf{p}^{(3)}(\omega_0)\cdot\mathbf{F}^{(a)}\big]\frac{\mathbf{F}^{(a)}}{|F^{(a)}|^2}$$

$$=\mathrm{Re}\{[\chi_{aa}(\omega_0)|F^{(a)}|^2$$

$$+\chi_{ab}(\omega_0)|F^{(b)}|^2]\cdot\mathbf{F}^{(a)}e^{-i\omega_0 t}\}, \quad (3.8)$$

where

$$\chi_{aa}(\omega_0)=-\tfrac{2}{5}|p_{12}|^4\alpha_{12}(\omega_0), \quad (3.8a)$$

$$\chi_{ab}(\omega_0)=-(2/15)|p_{12}|^4(1+2\cos^2\phi)\beta_{12}(\omega_0). \quad (3.8b)$$

Thus, for the added average absorbed power, in addition to that due to $\mathbf{p}^{(1)}(t)$, we take a time average of the scalar product of the rate of change of $\mathbf{p}_a^{(3)}(\omega_0)$ and the corresponding component of the radiation field together with the proper constant coefficient. It can easily be seen that part of this added absorbed power depends on ϕ through the ϕ dependence of $\chi_{ab}(\omega_0)$; explicitly, it is

$$-\frac{8\pi N}{15c}|p_{12}|^4|F^{(a)}|^2|F^{(b)}|^2\,\mathrm{Im}\beta_{12}(\omega_0)(\cos^2\phi)\omega_0, \quad (3.9a)$$

where c is the velocity of light. Similarly, it can be shown that the ϕ-dependent part of the absorbed power at $\omega_0+\Delta\omega$ is

$$-\frac{8\pi N}{15c}|p_{12}|^4|F^{(a)}|^2|F^{(b)}|^2$$

$$\times\mathrm{Im}a_{12}(\omega_0+\Delta\omega)(\cos^2\phi)(\omega_0+\Delta\omega). \quad (3.9b)$$

Thus, whether the absorbed power corresponding to Eqs. (3.9a) and (3.9b) is positive or negative will depend upon the signs of $\mathrm{Im}\beta_{12}(\omega_0)$ and $\mathrm{Im}a_{12}(\omega_0+\Delta\omega)$. It can be shown straightforwardly from Eqs. (3.7b), (3.7c), and (3.4) that, in general, they could each be positive or negative independently depending upon the values of the various parameters. However, this is not so in the particular ranges of parameters stipulated here: The Doppler linewidth, characterized by σ, is much greater than $\Delta\omega$, and $\Delta\omega$ is much greater than the collision linewidth or

$$\sigma\tau\gg\Delta\omega\tau\gg1. \quad (3.10)$$

Furthermore,

$$\omega_0\gg\Delta\omega. \quad (3.11)$$

The Doppler effect can now be taken into consideration by replacing ω_0 and $\omega_0+\Delta\omega$ by their Doppler-shifted values and then averaging over the Boltzmann distribution for the translational velocity, v, of the molecules. In performing the integration over v, by virtue of Eqs. (3.10) and (3.11), the integrands $\mathrm{Im}\beta_{12}$ and $\mathrm{Im}\alpha_{12}$ are rapidly varying functions of v, and the major net contributions of the integrals come from two points—where the two Doppler-shifted frequencies are close to ω_{12}. Hence, all the slowly varying factors of the integrands can be taken out of the integral signs and evaluated at these two points, leaving rather simple integrations to be performed. Thus, we obtain:

$$\mathrm{Im}\beta_{12}(\omega_0)=\frac{3\Delta\rho_{12}^{(0)}\omega_0\pi}{4\hbar^2\Delta\omega^2\sigma\sqrt{\pi}}>0, \quad (3.12a)$$

and

$$\mathrm{Im}\alpha_{12}(\omega_0+\Delta\omega)=\frac{3\Delta\rho_{12}^{(0)}(\omega_0+\Delta\omega)\pi}{4\hbar^2\Delta\omega^2\sigma\sqrt{\pi}}>0, \quad (3.12b)$$

since $\Delta\rho_{12}{}^{(0)}>0$, where $\sigma\sqrt{\pi}$ comes from the normalizing constant in the Boltzmann factor, and, by virtue of Eqs. (3.10) and (3.11), terms of the order $\Delta\omega/\omega_0$, $(\Delta\omega/\sigma)^2$, $(\Delta\omega\tau)^{-2}$ and smaller are neglected. On the basis of Eqs. (3.9a), (3.9b), (3.12a), and (3.12b), we reach the important conclusion: for maximum absorption,

$$\phi=\pm[(2n+1)/2]\pi, \quad n=0, 1, 2\cdots, \quad (3.13)$$

so that the added absorbed power corresponding to $\mathbf{p}^{(3)}(t)$, which is negative, due to the nonlinear effects considered here would be the minimum. In other words, for maximum absorption, the two linearly polarized fields $\mathbf{F}^{(a)}$ and $\mathbf{F}^{(b)}$ must be at right angles to each other.

The reason for this result is quantum mechanical in nature, but a simple physical explanation can perhaps be given in terms of the saturation effect. When the gas is irradiated by two linearly polarized oscillating fields, the resultant field is amplitude modulated at the difference frequency which is small compared with the oscillation frequencies. The maximum instantaneous magnitude of the resultant field depends upon the relative orientation of polarization of the two linearly polarized fields; nevertheless, without taking into account saturation, the absorptions of the two fields are completely independent. Consider now the effect of saturation. Saturation depends upon the instantaneous magnitude of the resultant field when the modulation is slow. When the two fields are perpendicular to each other, the maximum instantaneous magnitude of the resultant field is the least as compared with any other relative orientations; therefore, it does not drive as deeply into the saturation region. Consequently, maximum absorption is expected when the two fields are perpendicular to each other.

An effect similar to this should also be present in the magnetic resonance of an ensemble of spin-1/2 nuclei under the simultaneous influence of two linearly polarized rf fields of different frequencies but well within the inhomogeneously broadened absorption line. This could be shown by either using the density matrix formalism as done here or using the phenomenological equation of Bloch assuming that $T_1=T_2$ and the rf field consists of two frequency components.

IV. EXCITATION BY THREE OSCILLATING FIELDS AND PHASE RELATIONSHIPS

We consider now the same gas placed in a static magnetic field $\mathfrak{IC}$ along the z axis and assume the two levels of interest E_1 and E_2 have angular momenta J of 0 and 1, respectively. In a representation in which the total Hamiltonian H_1,

$$H_1=H_0-g\mu_0\mathbf{J}\cdot\mathfrak{IC}, \quad (4.1)$$

corresponding to the internal molecular energy and the Zeeman energy is diagonal, the E_2 level will be split up (Fig. 2). In Eq. (4.1), g is the spectroscopic splitting factor of the E_2 state and μ_0 is the Bohr
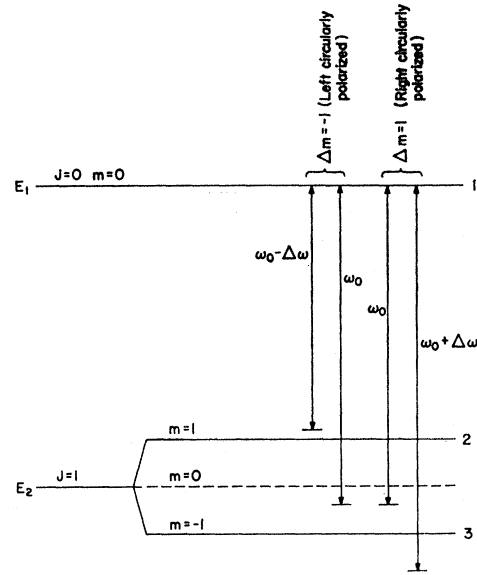

FIG. 2. Positions of Zeeman levels and the applied radiation.

magneton. Designating the levels of interest as level $1(E_1, J=0, m=0)$, $2(E_2, J=1, m=1)$, and $3(E_2, J=1, m=-1)$, the transition frequencies of interest are ω_{12}, ω_{23}, and ω_{13}. The applied radiation consists of two right circularly polarized fields at ω_0 and $\omega_0+\Delta\omega$, and two left circularly polarized fields at ω_0 and $\omega_0-\Delta\omega$ (Fig. 2)

$$F(t)=\mathrm{Re}[\mathbf{F}_+{}^{(1)}e^{-i(\omega_0+\Delta\omega)t-i\theta^+}+(\mathbf{F}_+{}^{(0)}+\mathbf{F}_-{}^{(0)})e^{-i\omega_0 t}$$
$$+\mathbf{F}_-{}^{(1)}e^{-i(\omega_0-\Delta\omega)t-i\theta^-}], \quad (4.2)$$

where in Eq. (4.2) $+$ and $-$ designate right and left circular polarizations, respectively. It is further assumed that

$$\omega_0\gg\Delta\omega, \omega_{23}, \quad (4.3a)$$

and no other transition frequencies, $\omega_{m'n'}$, are close to ω_0, ω_{12}, ω_{13} in the sense

$$|\omega_0-\omega_{12}| \approx |\omega_0-\omega_{13}|\ll|\omega_0-\omega_{m'n'}|. \quad (4.3b)$$

The perturbation term, $V(t)$, in the total Hamiltonian $H=H_1+V$ has the following matrix elements, neglecting the nonresonant counter-rotating terms:

$$V_{13}=V_{31}{}^*=\tfrac{1}{2}V_{+13}{}^{(0)}e^{-i\omega_0 t}+\tfrac{1}{2}V_{+13}{}^{(1)}e^{-i(\omega_0+\Delta\omega)t-i\theta^+},$$

$$V_{12}=V_{21}{}^*=\tfrac{1}{2}V_{-12}{}^{(0)}e^{-i\omega_0 t}+\tfrac{1}{2}V_{-12}{}^{(1)}e^{-i(\omega_0-\Delta\omega)t-i\theta^-}, \quad (4.4)$$

$$V_{23}=V_{32}=V_{11}=V_{22}=V_{33}=0,$$

where, with $\mathbf{p}$ as the dipole operator,

$$V_{\pm mn}{}^{(l)}\equiv-\langle m|\mathbf{p}\cdot\mathbf{F}_\pm{}^{(l)}|n\rangle, \quad l=0, 1. \quad (4.5)$$

The problem now is to determine the part of the power absorbed by the gas that depends upon the relative phases θ^+ and θ^- of the applied fields.

With the perturbation term V specified, Eq. (4.4), it is again possible to determine the induced dipole moments and the absorbed power at various frequencies

through a straightforward, though quite tedious, application of the routine outlined in Sec. II. The procedure is well illustrated in the problem treated in detail in Sec. III with the only essential difference in the fact that here no averaging over the orientation of the molecule is needed as such a degeneracy has been removed upon application of the static magnetic field.

In brief, starting with Eq. (4.4), we obtain through successive applications of Eqs. (2.7) and (2.9) all the matrix elements of $D^{(1)}$ and $D^{(2)}$. These in turn lead to the various Fourier components of all the elements of $D^{(3)}$ and, therefore, $p^{(3)}(t)$ via Eqs. (2.9) and (2.3). Since the eventual purpose is to compute the absorption of each component of the radiation fields, of particular interest are: The component of $\mathbf{p}^{(3)}(t)$ varying harmonically at the angular frequency $\omega_0+\Delta\omega$ with the phase θ^+ that is right circularly polarized, the components varying at ω_0 that are either right or left circularly polarized, and the component varying at $\omega_0-\Delta\omega$ with the phase θ^- that is left circularly polarized. Furthermore, we are only interested in those terms in the amplitude of each of these components which depend upon the relative phases θ^+ and θ^-. These observations simply the problem substantially, since of the multitude of terms in $\mathbf{p}^{(3)}(t)$ only a few terms meet all these requirements. Take, for example, the $\omega_0+\Delta\omega$ component. The terms of interest come entirely from $D_{13}^{(3)}(\omega_0+\Delta\omega)$; they are

$$\mathbf{p}_+^{(3)}(\omega_0+\Delta\omega;\theta^+,\theta^-)$$
$$=2\mathrm{Re}[\mathbf{p}_{31}D_{13}^{(3)}(\omega_0+\Delta\omega;\theta^+,\theta^-)]$$
$$=\mathrm{Re}[\mathcal{X}_+(\omega_0+\Delta\omega)e^{i(\theta^++\theta^-)}|F_-^{(0)}|\,|F_1^{(1)}|\,\mathbf{F}_+^{(0)}$$
$$\times e^{-i(\omega_0+\Delta\omega)t-i\theta^+}], \quad (4.6)$$

where

$$\mathcal{X}_+(\omega_0+\Delta\omega)=-\frac{i|p_{13}|^2|p_{12}|^2(\omega_0+\Delta\omega)\tau^3}{4\hbar^2(1-i\Omega_{13}^{(+)}\tau)}$$
$$\times\left[\Delta\rho_{12}^{(0)}\left(\frac{1}{1+i\Omega_{12}\tau}+\frac{1}{1-i\Omega_{12}^{(-)}\tau}\right.\right.$$
$$\left.+\frac{1}{(1+i\Omega_{23}\tau)(1-i\Omega_{12}^{(-)}\tau)}\right)$$
$$\left.+\Delta\rho_{13}^{(0)}\frac{1}{(1+i\Omega_{23}\tau)(1+i\Omega_{13}\tau)}\right], \quad (4.6a)$$

and

$$\Omega_{13}=\omega_{13}-\omega_0>0, \quad \Omega_{13}^{(+)}=\omega_{13}-(\omega_0+\Delta\omega)<0,$$
$$\Omega_{12}=\omega_{12}-\omega_0<0, \quad \Omega_{12}^{(-)}=\omega_{12}-(\omega_0-\Delta\omega)>0, \quad (4.7)$$
$$\Omega_{23}=\omega_{23}-\Delta\omega.$$

The corresponding absorbed power is found using a procedure similar to that used in Sec. III. The only difference is that in performing the integration over the translational velocity of the molecule in order to take into account the Doppler effect, major contributions

come from four points—where the Doppler shifted frequencies $\omega_0^{(d)}$ and $(\omega_0+\Delta\omega)^{(d)}$ are near ω_{13}, and $\omega_0^{(d)}$ and $(\omega_0-\Delta\omega)^{(d)}$ are near ω_{12}. Carrying out the integrations, we obtain for the absorbed power at $\omega_0+\Delta\omega$

$$[(\omega_0+\Delta\omega)2\pi N/c]|F_+^{(0)}|\,|F_+^{(1)}|F_-^{(0)}|\,|F_-^{(1)}|$$
$$\times|\mathcal{X}_+(\omega_0+\Delta\omega)|\,\sin(\phi_+^{(1)}+\theta^++\theta^-), \quad (4.8)$$

where

$$\phi_+^{(1)}=\tan^{-1}\left(\frac{\mathrm{Im}\mathcal{X}_+(\omega_0+\Delta\omega)}{\mathrm{Re}\mathcal{X}_+(\omega_0+\Delta\omega)}\right), \quad (4.8a)$$

and

$$\mathrm{Im}\mathcal{X}_+(\omega_0+\Delta\omega)=-\frac{|p_{13}|^2|p_{12}|^2(\omega_0+\Delta\omega)\tau^2\pi}{4\hbar^2\sigma\sqrt{\pi}}$$
$$\times\left(\frac{2}{1+\Omega_{23}^2\tau^2}+\frac{1}{1+(\Omega_{23}-\Delta\omega)^2\tau^2}\right),$$

$$\mathrm{Re}\mathcal{X}_+(\omega_0+\Delta\omega)=-\frac{|p_{13}|^2|p_{12}|^2(\omega_0+\Delta\omega)\tau^2\pi}{4\hbar^2\sigma\sqrt{\pi}}$$
$$\times\left(\frac{1}{\Delta\omega\tau}+\frac{(\Delta\omega-\Omega_{23})\tau}{1+(\Omega_{23}-\Delta\omega)^2\tau^2}\right). \quad (4.8b)$$

In Eqs. (4.8a) and (4.8b), again, terms with contributions of the order $(\Delta\omega/\sigma)^2$, $\Delta\omega/\omega_0$, $(\Delta\omega\tau)^{-2}$ and smaller are ignored and use is made of the fact that $\Delta\rho_{12}^{(0)}=\Delta\rho_{13}^{(0)}+O(\omega_{23}/\omega_0)$ by virtue of Eqs. (2.5) and (2.8). Similar computations can be carried out for the remaining components of the radiation. Omitting all details, the power absorbed of the right circularly polarized field at ω_0 that depends upon $\theta^++\theta^-$ is:

$$(\omega_0 2\pi N/c)|F_+^{(0)}|\,|F_+^{(1)}|\,|F_-^{(0)}|\,|F_-^{(1)}|$$
$$\times|\mathcal{X}_+(\omega_0)|\,\sin(\phi_+^{(0)}-\theta^+-\theta^-), \quad (4.9)$$

where

$$\phi_+^{(0)}=\tan^{-1}\left(\frac{\mathrm{Im}\mathcal{X}_+(\omega_0)}{\mathrm{Re}\mathcal{X}_+(\omega_0)}\right), \quad (4.9a)$$

and

$$\mathrm{Im}\mathcal{X}_+(\omega_0)=-\frac{|p_{13}|^2|p_{12}|^2\omega_0\tau^2\pi}{4\hbar^2\sigma\sqrt{\pi}}$$
$$\times\left(\frac{2}{1+\Omega_{23}^2\tau^2}+\frac{1}{1+(\Omega_{23}+\Delta\omega)^2\tau^2}\right),$$

$$\mathrm{Re}\mathcal{X}_+(\omega_0)=\frac{|p_{13}|^2|p_{12}|^2\omega_0\tau^2\pi}{4\hbar^2\sigma\sqrt{\pi}}$$
$$\times\left(\frac{1}{\Delta\omega\tau}+\frac{(\Omega_{23}+\Delta\omega)\tau}{1+(\Omega_{23}+\Delta\omega)^2\tau^2}\right). \quad (4.9b)$$

The absorbed power for the two left circularly polarized fields at ω_0 and $\omega_0-\Delta\omega$ can be obtained from the absorbed power for the two right circularly polarized fields at ω_0 and $\omega_0+\Delta\omega$, respectively, with the simple replacements:

$$\Delta\omega\longrightarrow-\Delta\omega,$$

and for the indices:

$$(+) \leftrightarrow (-) \quad \text{and} \quad 2 \leftrightarrow 3$$

by virtue of the inherent symmetry of the problem. It is then easy to show that the power absorbed from the left circularly polarized field at ω_0 that depends upon θ^+ and θ^- is

$$(\omega_0 2\pi N/c)|F_+^{(0)}||F_+^{(1)}||F_-^{(0)}||F_-^{(1)}|$$
$$\times |X_-(\omega_0)|\sin(\phi_-^{(0)} - \theta^+ - \theta^-), \quad (4.10)$$

where

$$\phi_-^{(0)} = \pi - \phi_+^{(0)},$$
$$|X_-(\omega_0)| = |X_+(\omega_0)|, \quad (4.10a)$$

and the power absorbed at $\omega_0 - \Delta\omega$ that depends upon θ^+ and θ^- is

$$[(\omega_0 - \Delta\omega)2\pi N/c]|F_+^{(0)}||F_+^{(1)}||F_-^{(0)}||F_-^{(1)}|$$
$$\times |X_-(\omega_0 - \Delta\omega)| \sin(\phi_-^{(1)} + \theta^+ + \theta^-), \quad (4.11)$$

where

$$\phi_-^{(1)} = \pi - \phi_+^{(1)},$$
$$|X_-(\omega_0 - \Delta\omega)| = |X_+(\omega_0 + \Delta\omega)|. \quad (4.11a)$$

Equations (4.8)–(4.11) give the necessary relations to determine when absorption of each component is at a maximum; in general, it is not possible to find one $\theta^+ + \theta^-$ which would give maximum absorption of all four components of the applied field. However, in the near resonance range in the sense:

$$\Omega_{23}^2 \ll \Delta\omega\tau^{-1}, \quad \text{or} \quad \omega_{23} \approx \Delta\omega. \quad (4.12)$$

It is clear from Eqs. (4.8)–(4.11) that

$$\phi_+^{(1)} \cong \phi_+^{(0)} \cong \phi_-^{(1)} \cong \phi_-^{(0)} \cong -\pi/2. \quad (4.13)$$

It follows unambiguously from Eq. (4.13) and Eqs. (4.8)–(4.11) that for maximum absorption of all four components, one must have, in the case Eq. (4.12) holds,

$$\theta^+ + \theta^- = \pi \pm 2n\pi, \quad n = 0, 1, 2 \cdots. \quad (4.14)$$

The physical reason for this result is similar to that of the case studied in Sec. III.

A detailed discussion of the application of the results obtained in these sections to gaseous optical masers is given in the subsequent section.

V. APPLICATION TO GASEOUS OPTICAL MASERS

We now consider the role of the nonlinearity due to saturation and coherence effects in determining the relationships among the longitudinal modes in a gaseous optical maser. In such a maser, such a nonlinearity must come into play, since the amplitude of the maser output is essentially determined by such nonlinearity as that due to saturation. In attempting to apply directly the results obtained previously for a normal gas

to the case where a population inversion and coherent stimulated emission occur, however, the basic assumption is used that the method of calculation and hence the results obtained are equally valid in the presence of a negative temperature[16] characterized by the population inversion maintained by the pumping process. The relaxation time, τ, is now the characteristic time constant with which the gas, upon perturbation, returns to its normal population inversion with the pumping process on; or, τ is the reciprocal natural linewidth of the stimulated emission due to transitions between the inverted states. In considering the Doppler effect which involves the translational velocity of the molecules, the normal thermodynamic temperature of the gas can still be used. Accepting these usual assumptions, with minor modifications, the two absorption problems considered in the previous sections apply directly to gaseous masers.

Consider first a normal gaseous He-Ne optical maser in the absence of any appreciable longitudinal magnetic field. The strongest maser line[2,17] corresponds to the Ne transition $2s_2 \to 2p_4$ (Paschen notation) at 1.1523 μ. We estimate that the Doppler width (σ) of this emission line in the absence of maser action is roughly 1 kMc/sec and τ is of the order of 10^{-7} sec at 1 mm Hg pressure He and 0.1 mm Hg pressure Ne. For a 1-m long maser, the separation in frequency ($\Delta\omega$) between the longitudinal modes of the Fabry-Perot cavity is 150 Mc/sec. Thus, conditions (3.10) and (3.11) of Sec. III are satisfied. Assume that the maser excitation is strong enough so that it can oscillate simultaneously in two cavity modes separated by 150 Mc/sec. Normally, the maser output in neither mode is polarized. However, in the event there is slight asymmetry in the maser or in the presence of a very weak transverse magnetic field, the maser output in each mode is linearly polarized. The polarizations are observed[6] to be perpendicular to each other. On the basis of the computations made in Sec. III, it is clear that, in the presence of a population inversion between levels 1 and 2 (levels $2s_2$ and $2p_4$), $\Delta\rho_{12}^{(0)} < 0$ and maximum negative absorption or maser gain for both longitudinal modes occurs when the two linearly polarizations are perpendicular to each other. Consequently, oscillations corresponding to this relative orientation get built up most rapidly and eventually prevail.

It should be pointed out that not only the absorptive part but also the dispersive part of the dipole moment contains a small nonlinear term which depends upon the relative orientation of polarization of the two fields. This leads to a small shift in the maser oscillation frequencies. However, due to the fact that the maser emission lines are well within the Doppler-broadened

[16] See, for example, A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, New York, 1961), pp. 89–92, 134 ff.

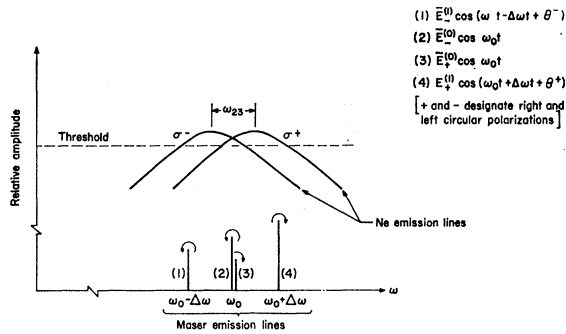[17] G. F. Koster and H. Statz, J. Appl. Phys. **32**, 2054 (1961).

FIG. 3. For suitable magnetic fields, the maser could oscillate in at least three modes simultaneously. Experimental evidence shows that $\theta^+ + \theta^- = \pi$, when $\omega_{23} \cong \Delta\omega$.

Ne emission line, the change in the negative absorption due to this small shift in frequency is quite small. For the particular case considered here, it is about 2 orders of magnitude smaller than that due to the nonlinear absorptive part of the induced dipole moment which depends upon the relative orientation of polarization of the fields.

In the presence of an applied longitudinal magnetic field along the maser axis, the $2s_2$ and $2p_4$ levels with angular momenta of $J=1$ and $J=2$, respectively, will consequently be split up into 3 and 5 levels each. Since the spectroscopic splitting factor g for the $2s_2$ and $2p_4$ levels are roughly the same,[18] the problem is qualitatively not very different from the mathematically much simpler case of $J=0 \to J=1$ as considered in Sec. IV. Thus the energy schematic depicted in Fig. 2 may be considered as a reasonable model for the present problem; and, of course, here $\Delta\rho_{12}^{(0)} \approx \Delta\rho_{13}^{(0)} < 0$ and the absorption is negative. Assume now the excitation is strong enough so that, as shown in Fig. 3, for certain magnetic field strength oscillations of right and left circularly polarized fields can take place in the ω_0 and $\omega_0 + \Delta\omega$, and ω_0 and $\omega_0 - \Delta\omega$ modes, respectively. The question is then what is the phase relationship among these modes. It is clear that according to the present theory, we again look for that phase relationship among the waves whereby the negative absorption, or the gain of the gaseous medium, of all the waves is at a maximum. This was done in Sec. IV.

Paananen[7] has observed, by directly monitoring, the 150-Mc/sec beat output of a phototube, used to measure the maser output, as a function of the applied longitudinal magnetic field (Fig. 4). Apart from the sharp dip at $\omega_{23} \cong \Delta\omega$ (where ω_{23} is twice the Zeeman frequency), the remaining feature is easily understood. At zero magnetic field, one of the cavity modes is situated somewhere near the center of the $2s_2 \to 2p_4$ emission line. If the maser excitation is only moderate, only one mode at ω_0 can oscillate. As the magnetic field is increased, the two Zeeman components ($\Delta m = \pm 1$, corre-

sponding to transitions: level $1 \to 3$ and $1 \to 2$ in Fig. 2) of the emission lines move to positions more symmetric with respect to the cavity modes so that right circularly polarized fields ($\Delta m = +1$) may oscillate in both ω_0 and $\omega_0 + \Delta\omega$ modes, similarly with the left circularly polarized fields ($\Delta m = -1$) at ω_0 and $\omega_0 - \Delta\omega$. As all four waves fall on the photocathode, the response of which is proportional to the square of the total incident field, the two right circularly polarized fields mix to give a 150-Mc/sec output and likewise the two left circularly polarized fields. When, and only when, these two 150-Mc/sec output signals have equal amplitude and opposite phases, they cancel completely; otherwise there is always a 150-Mc/sec output when the maser oscillates in all these modes. The dip at $\omega_{23} = \Delta\omega$ implies a particular phase relationship among the various modes of the maser output at $\omega_0 - \Delta\omega$, ω_0 and $\omega_0 + \Delta\omega$. It can be shown straightforwardly by taking the square of the assumed maser output, as shown in Fig. 3, that the amplitudes of the two 150-Mc/sec components are always equal when $\omega_{23} = \Delta\omega$ and they have opposite phases near $\omega_{23} = \Delta\omega$ only when $\theta^- + \theta^+ = \pi$. This is precisely the phase relationship found theoretically on the basis of the result obtained in Sec. IV, Eq. (4.14). Furthermore, if ω_0 is not exactly halfway between ω_{12} and ω_{13}, complete cancellation of the two 150-Mc/sec components is expected on the basis of the computations given in Sec. IV to be possible only at $\omega_{23} = \Delta\omega$ with a sharp null as observed (Fig. 4). It is important to note that the sum of the relative phases $(\theta^+ + \theta^-)$ is spatially independent. This is because the spatially dependent phase shifts of the $\omega_0 + \Delta\omega$ and $\omega_0 - \Delta\omega$ modes are symmetric with respect to the spatially dependent phase shift of the ω_0 mode; consequently, their sum is always nil. The effects discussed in this paper are not altered if the small amount of pulling[18] in all the components are taken into account. When the condition (4.12) does not hold, it is not clear to us what the phase
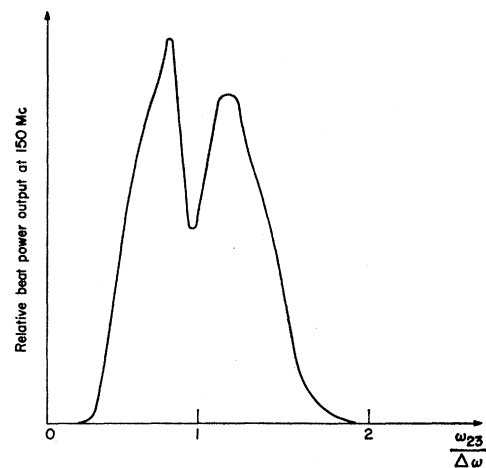

FIG. 4. The 150-Mc/sec beat note as a function of $\omega_{23}/\Delta\omega$.

[18] H. Statz, R. A. Paananen, and G. F. Koster, J. Appl. Phys. 32, 2319 (1962).

relationship among the modes should be, since it is not possible to meet the simple unambiguous criterion that the negative absorption of all modes shall be maximum simultaneously. This question is still under study.

Magnetoacoustic Measurements in the Noble Metals at 350 Mc/sec*

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Further magnetoacoustic measurements on the noble metals are presented for frequencies up to 350 Mc/sec. Plots of the ultrasonic pulse height, h, vs the reciprocal of the magnetic field strength, $1/H$, show 20 or more maxima and minima for several orientations in each metal. Fermi surface dimensions are calculated from the periods in $1/H$ of the magnetoacoustic oscillations using the interpretation that the periodicity arises from those portions of the Fermi surface which are extremal in the $q \times H$ direction of momentum space; Fermi surface cross sections viewed along the [100], [110], [111], and [112] directions are shown in detail. The results are compared with Fermi surface dimensions given by:(1) other magnetoacoustic effect data, (2) de Haas-van Alphen and anomalous skin-effect data, and (3) recent band theory calculations. Some simple calculations of electron mean free paths and collision relaxation times are given.

INTRODUCTION

IN this paper we shall present further noble metals Fermi surface dimensions[1-6] derived from magneto-acoustic-effect data. The oscillatory variation of ultrasonic attenuation as a function of magnetic field intensity (magnetoacoustic effect) occurs only in very pure metals at low temperatures due to the requirement that the electronic mean free path be much greater than the sound wavelength, λ. The oscillations which are periodic in H^{-1} (H is the magnetic field strength) occur because of geometrical coincidences between certain electron orbits and the periodic electric fields accompanying the sound wave. The effect, when **H** is perpendicular to the sound propagation vector **q** (q is $2\pi/\lambda$), gives the dimensions and ultimately the shape of the Fermi surface if we make the interpretation that the period observed in H^{-1} is determined by the Fermi momentum perpendicular to **H** and **q** at those portions of the surface where this momentum is extremal.[7-10]

If $\hbar k$ is such a momentum and $\Delta(H^{-1})$ is the corresponding magnetoacoustic period, then

$$\hbar k = (e\lambda/2c)[1/\Delta(1/H)].\qquad (1)$$

Almost all data were taken using longitudinal sound waves, the sound propagation directions studied being [100], [110], and [111] for all three metals, and additionally [112] in gold and copper. In these experiments the magnetic field can be given any direction in a plane perpendicular to **q**. The longitudinal velocities, lattice constants, and free-electron sphere radii for the noble metals are given in Table I.[11,12] The experimental techniques and preparation of samples were essentially the same as those discussed in an earlier article[5] except that we now have equipment, constructed in our laboratory, which increases our frequency range to 350 Mc/sec. All three metals were of nominal purity 99.999% or better.

The method of plotting and analyzing the data was the same as described earlier.[5] The extremal Fermi surface dimensions corresponding to the observed periods of oscillation are interpreted in terms of the Fermi surface model shown in the extended zone scheme in Fig. 1. Some of the extremal orbits which we shall discuss are also shown in the figure.

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