The dependence of the gap width on temperature has been estimated from the differential conductance at zero bias voltage. Good agreement is found with the predicted BCS variation if the BCS curve is scaled by a constant multiplier to give the best visual fit to the data.

Structure, thought to indicate peaks in the phonon spectrum of mercury, has been observed in the first and second derivatives of the tunneling characteristic. $d^2I/dV^2$ exhibits two groups of minima, one group at low energies, in which the peaks are relatively large and sharp, and a second group at higher energies, in which the structure is small and broad. Neutron scattering experiments performed on mercury might provide further information about these peaks.

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# Double-Resonance Phenomena in the Gaseous Laser*

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The theory of double-resonance phenomena, or the simultaneous action of optical transitions and rf perturbations between the Zeeman sublevels of an atom, is extended to the induced emission and absorption processes effected by the monochromatic radiation and the discrete resonances involved in the gas laser. Expressions for the probability amplitudes of the Zeeman sublevels under such conditions are derived from time-dependent perturbation theory, the lifetimes of the states being introduced in a phenomenological way. Zeeman splittings of both upper and lower states involved in the laser transition are considered, and an effective term diagram used to discuss the results. Small signal approximations are applied to consider some specific cases, which show that the light beats seen in spontaneous emission when the rf frequency $\omega_0$ equals the Larmor frequency, and in other instances, do not occur in induced emission unless such frequencies coincide with axial resonances of the laser cavity. Variations in the intensity and resonance effects are indicated at Zeeman splittings corresponding to $\omega_0$, with similar variations in the amplitude of any beat frequencies due to multiple axial resonances. Additional coherence effects occur where levels cross, which will give rise to changes in the intensity and polarization of the induced emission at the corresponding frequency.

## 1. INTRODUCTION

The use of double-resonance phenomena for the investigation of atomic structure was first discussed by Bitter[1] with reference to measurements on the hyperfine structure of excited atomic states. Some clarification of the original ideas was also given later by Pryce.[2] In double-resonance experiments, atoms are subjected simultaneously to radiations at both optical and radio frequencies, which are near to the atomic resonant frequencies involved in electric dipole and magnetic dipole transitions, respectively. Experiments on the effect of such rf perturbations between the Zeeman sublevels of an excited state were proposed by Brossel and Kastler,[3] and the first experiment on such double-resonance phenomena was done by Brossel and Bitter[4] using mercury vapor situated in a uniform magnetic field. Here suitable optical resonance radiation excited the mercury atoms from the ground state $^1S_0$ to the state $m=0$ of the excited state $^3P_1$, the wavelength being 2537 Å. Transitions to the states $m=\pm1$ were then induced by an rf magnetic field oscillating perpendicularly to the dc magnetic field at the Larmor frequency. These rf transitions were detected by the changes they produced on the intensity and polarization of the spontaneously emitted optical radiation. Resonance curves showing the variation of light intensity with dc magnetic field at a given rf frequency were obtained. From these curves quite accurate values of the $g$ factor of the excited state were deduced, and the width of such resonances at low rf field amplitudes represented a measure of the natural linewidth of the excited state.

Dodd *et al.*[5] later did similar experiments which showed that the fluorescent light in such a double-resonance experiment is strongly modulated at the Larmor frequency and at multiples of this, depending on the dc magnetic field and on the orientations of the incident and observed polarizations of the optical radiation. Such modulation effects are due to beats between the spontaneously emitted radiations at

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different frequencies from the excited Zeeman sublevels to the ground state, which have been made coherent by the rf field inducing transitions between them. Similar variations in the intensity of the fluorescent light were observed in the level crossing experiments on helium carried out by Colgrove *et al.*[6] Here the coherence is imparted to the overlapping Zeeman levels by suitable resonance transitions from a common lower state.

Dodd and Series[7] have given an extensive theoretical treatment of double-resonance phenomena. This is applicable to the excitation by resonance radiation and subsequent decay by spontaneous emission of optical radiation from the Zeeman sublevels of an excited state with $J=1$ to a ground state with $J=0$. It assumes that the excitation rate, or optical perturbation, is small compared with the radio frequency perturbation which induces transitions between the Zeeman sublevels of the excited state. Coherence between these sublevels is thus imparted by this rf field, or by the excitation process when levels overlap, or by both of these processes. The theory cannot be derived in terms of the populations of the various states, but must consider that these are no longer independent when the rf field is applied. The phases of the probability amplitudes of the Zeeman sublevels are related to that of the rf field, and hence to each other. The fluorescent radiation from the various levels of a single atom then manifests this coherence on detection as light beats, which are due to interference between the optical radiations at frequencies separated by the Larmor frequency or multiples of this. Barrat, utilizing his elegant work on the theory of light trapping,[8] has formulated the same problem in terms of the density matrix[9] with essentially the same results. Here, the coherence between the Zeeman sublevels is apparent in the finite values of the off-diagonal elements of the density matrix. Later experimental work on double resonance[10,11] phenomena in resonance radiation has amply confirmed the predictions of the theory. In passing, we may note that the absorption counterpart of these experiments is the work of Bell and Bloom[12] on an atomic vapor subjected to both suitably polarized optical radiation and rf fields acting on the Zeeman sublevels of the ground state. In this experiment, which was suggested by Dehmelt, a phenomenological treatment based on the Bloch equations[13] was used to explain the phenomena. It is possible that a similar application of these equations could be made to discuss double-resonance phenomena involving excited states.

In the present account the work on double resonance, and in particular that described in Refs. 7, 8, and 9, is extended to conditions applicable in the gas laser. Here, in contrast to the spontaneous emission processes considered above, we must now deal with the induced emission due to the highly monochromatic nature and discrete frequencies involved in the laser radiation. The characteristics and resonant frequencies of the optical resonator must be considered, and laser oscillations of the particular polarization and frequency corresponding to a given optical transition must be possible before such a transition will be induced to emit. An example of a resonator which is isotropic as regards polarization is that used in the internal optics laser, which puts no polarization constraint on the interaction of radiation with the excited atoms within the resonator. Apart from this requirement, such a resonator may be made planar, spherical, or confocal in the usual way. A polarization constrained resonator would be obtained when Brewster angle windows are interposed inside the cavity, and is analogous to the use of an analyzer inserted in the output radiation from spontaneous emission processes in double resonance work. The effect of this can be deduced from the general results to be derived. The treatment will consider the coherence and radiative processes applicable to a single atom concerned in the laser emission. This may be regarded as the miscroscopic system from which the macroscopic properties of the ensemble of atoms such as polarization and intensity variations are derived. Excitation of the atoms occurs either by collisions with other excited metastable atoms or by preferential electronic interaction with the different atomic states. We shall assume that such processes are isotropic for the various Zeeman sublevels and, in general, will only consider an atom in a particular atomic state at a time $t=0$. General time-dependent perturbation equations involving both electric dipole and magnetic dipole matrix elements are derived which are valid for any level of the perturbations. To obtain some physical interpretation of the complicated equations the initial inducing optical perturbation is assumed small; the population of the excited levels is then determined by their decay time to lower lying atomic levels. The results so deduced thus correspond to a small signal theory. They are, however, valid for all ranges of dc magnetic field and for any level of the rf perturbation which establishes coherence between the Zeeman sublevels. Finally, some applications of the results to specific situations are considered.

## 2. THEORY

We assume initially that the laser operates between an upper level of total angular momentum $j=1$ and a

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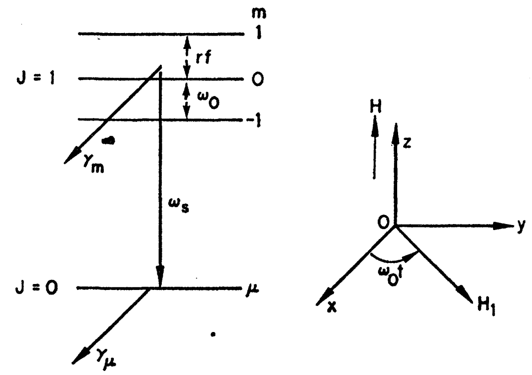


FIG. 1. Double-resonance arrangement considered.

lower level with $j=0$, and that there is no nuclear spin. There are thus three magnetic sublevels $|m\rangle$ in the excited state which have equal decay constants $\gamma_m$ to lower lying atomic levels. The lower level $|\mu\rangle$ does not split in the magnetic field and has a decay constant $\gamma_\mu$ to lower levels. Figure 1 shows the arrangement to be considered with the dc magnetic field acting along the $z$ axis, and the rf field along the $x$ axis, and rotating in the $xy$ plane. Magnetic dipole transitions are induced between the $|m\rangle$ levels by the rf magnetic field, and electric dipole transitions are induced between the $|m\rangle$ and $|\mu\rangle$ levels by the electric field in the laser. The results can be generalized later to more complicated situations such as occur in the He-Ne laser, where $j=1$ and $j=2$ for the upper and lower levels, respectively.

The complete Hamiltonian for the system is given by

$$\mathcal{H}=\mathcal{H}_0+\gamma\mathbf{J}\cdot\mathbf{H}+\mathcal{H}_{\mathrm{rf}}+\mathcal{H}_e+\mathcal{H}_D\,, \tag{1}$$

where $\gamma=ge/2mc$ is the gyromagnetic ratio, and $\mathcal{H}_0$ represents the unperturbed Hamiltonian. $\mathbf{J}\cdot\mathbf{H}$ is the perturbation due to the dc magnetic field, which is of magnitude $H$ along the $z$ axis, and $\mathcal{H}_{\mathrm{rf}}$ is the perturbation due to the rf magnetic field, which we consider to rotate with angular frequency $\omega_0$ about the $z$ axis in the $xy$ plane. $\mathcal{H}_D$ is the damping term introduced in a phenomenological way,[14] and which is diagonal in $\gamma_m$ and $\gamma_\mu$, and $\mathcal{H}_e$ is the electric dipole perturbation due to the laser emission. Using the electric dipole approximation, this is given by

$$\mathcal{H}_e=e\mathbf{E}\cdot\mathbf{r}\cos\omega_s t\,, \tag{2}$$

where $\mathbf{E}$ is the amplitude in magnitude and direction, and $\omega_s$ is the angular frequency of the laser radiation. The resulting time-dependent perturbation equation will be solved by regarding $\mathcal{H}_e$ as a perturbation on the eigenstates of the remaining part of the Hamiltonian. We thus write

$$\mathcal{H}=\mathcal{H}_1+\mathcal{H}_e \tag{3}$$

and proceed to find the eigenstates and eigenvalues of $\mathcal{H}_1$, where

$$\mathcal{H}_1=\mathcal{H}_0+\gamma\mathbf{J}\cdot\mathbf{H}+\mathcal{H}_{\mathrm{rf}}+\mathcal{H}_D\,. \tag{4}$$

We have

$$\begin{aligned}\mathcal{H}_{\mathrm{rf}}&=\gamma\mathbf{J}\cdot[(H_1\cos\omega_0 t)\mathbf{i}+(H_1\sin\omega_0 t)\mathbf{j}]\\&=\tfrac{1}{2}\gamma H_1[J_+e^{-i\omega_0 t}+J_-e^{i\omega_0 t}]\,,\end{aligned} \tag{5}$$

where $J\pm=(J_x\pm iJ_y)$. The Hamiltonian $\mathcal{H}_1$ may then be written as[15]

$$\mathcal{H}_1=e^{-(i/\hbar)J_z\omega_0 t}[\mathcal{H}_0+\gamma J_zH+\gamma H_1J_x+\mathcal{H}_D]e^{(i/\hbar)J_z\omega_0 t}\,, \tag{6}$$

and the time dependence of a state $|\,\rangle$ in the $\alpha jm$ representation given by

$$|\;\rangle=\textstyle\sum_m a_m(t)|m\rangle+a_\mu(t)|\mu\rangle\,, \tag{7}$$

is determined by the Schrödinger equation

$$i\hbar d|\;\rangle/dt=\mathcal{H}_1|\;\rangle\,. \tag{8}$$

As in other problems involving rf transitions between fine structure levels, it is convenient to apply the unitary transformation[16]

$$|'\rangle=e^{(i/\hbar)J_z\omega_0 t}|\;\rangle \tag{9}$$

to the equation of motion (8). Referring to Fig. 2, this now becomes

$$i\hbar d|'\rangle/dt=(\mathcal{H}_1-\omega_0 J_z)|'\rangle=\mathcal{H}'|'\rangle\,, \tag{10}$$

where the Hamiltonian $\mathcal{H}'$ in the coordinate system $x'y'z$ rotating about $Oz$ with angular velocity $\omega_0$ is now independent of time. Expanding the state $|'\rangle$ in the form

$$|'\rangle=\textstyle\sum_m a_m'|m\rangle+a_\mu'|\mu\rangle \tag{11}$$

and using Eqs. (7) and (9), we find that

$$a_m'=e^{im\omega_0 t}a_m\,,\quad a_\mu'=a_\mu(J_\mu=0)\,, \tag{12}$$

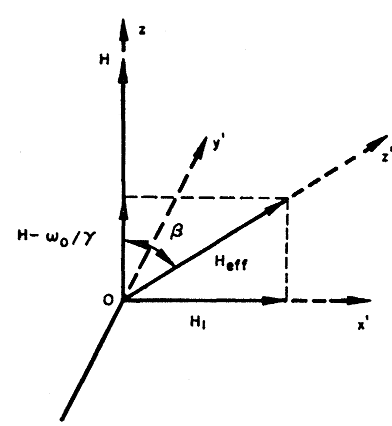


FIG. 2. Effective magnetic field and axis of quantization $z'$ in the rotating coordinate system.

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[16] I. I. Rabi, N. F. Ramsey, and J. Schwinger, Rev. Mod. Phys. **26**, 167 (1954).

and also that the effective electric dipole perturbation matrix element in the rotating coordinate system must be taken as

$$e^{im\omega_0 t}\langle m|\mathcal{H}_e|\mu\rangle a_\mu \quad \text{or} \quad e^{im\omega_0 t}\langle m|\mathcal{H}_e|\mu\rangle e^{-i\mu\omega_0 t}a_\mu\,, \qquad (13)$$

etc., for the cases where the $J$ value of the lower laser level is zero or finite. Equation (13) follows by applying the above unitary transformation to the operator $\mathcal{H}_e$, which gives

$$\mathcal{H}_e'=e^{(i/\hbar)J_z\omega_0 t}\mathcal{H}_e e^{-(i/\hbar)J_z\omega_0 t}. \qquad (14)$$

From Eq. (10) the Hamiltonian $\mathcal{H}'$, which is now independent of time, may be written as

$$\mathcal{H}'=\mathcal{H}_0+\mathcal{H}_D+\delta J_z+bJ_x\,, \qquad (15)$$

where $\delta=\omega-\omega_0$, $\omega=\gamma H$, and $b=\gamma H_1$. For the desired representation, we require the eigenstates and eigenvalues of the Hamiltonian $\mathcal{H}'$. To obtain these we apply a unitary transformation $U$ which rotates the coordinate system through an angle $\beta$ about the $Oy'$ axis, where $\tan\beta=b/\delta$ as shown in Fig. 2. The axis of quantization is then taken along $Oz'$ the direction of $H_{\rm eff}$, the effective magnetic field in the rotating coordinate system. Since $\mathcal{H}_0$, and $\mathcal{H}_D$ commute with this unitary operator they are unchanged. Hence, resolving $J_z$ and $J_x$ along the effective field direction we obtain

$$\mathcal{H}'=\mathcal{H}_0+\mathcal{H}_D+pJ_z'\,, \qquad (16)$$

where $p=(\delta^2+b^2)^{1/2}$, and $J_z'$ is the component of total angular momentum along the new axis of quantization.

The perturbation treatment is now carried out in this representation, in which the eigenstates and eigenvalues are given by

$$|M\rangle=|\alpha jJ_z'=M\hbar\rangle\,, \quad \text{and} \quad E(\alpha,j)+pM\hbar\,, \qquad (17)$$

respectively, and where $M=-J\cdots+J$. Eigenstates $|M\rangle$ are linear combinations of the eigenstates $|m\rangle$ and may be obtained by a rotation of the coordinate system $Ox'y'z$ through an angle $\beta$ about the $y'$ axis. This rotation is effected by the irreducible representation of the rotation group corresponding to the angular momentum $j$, and which may be written as[17]

$$|M\rangle=\sum_m\langle m|M\rangle|m\rangle=\mathfrak{D}^j(O,\beta,O)_{mM}|m\rangle\,, \qquad (18)$$

where

$$\mathfrak{D}_{\mu'\mu}{}^j=\sum_K\frac{(-1)^K[(j+\mu)!(j-\mu)!(j+\mu')!(j-\mu')!]^{1/2}}{(j-\mu'-K)!(j+\mu-K)!K!(K+\mu'-\mu)!}\times\cos^u\tfrac{1}{2}\beta\,\sin^v\tfrac{1}{2}\beta\,. \qquad (19)$$

$u=2j+\mu-\mu'-2K$, $v=2K+\mu'-\mu$, and the summations over $K$ are taken over all integers in the range between the larger of the numbers $O$ or $\mu-\mu'$ and the smaller of the numbers $j-\mu'$ and $j+\mu$. Thus, for $j=1$ we obtain from Eq. (19) the result

$$\langle m|M\rangle=\begin{array}{r|ccc|} & -1 & 0 & 1\\ \hline -1 & c^2\psi & -\sqrt{2}s\psi c\psi & s^2\psi\\ 0 & \sqrt{2}s\psi c\psi & c^2\psi-s^2\psi & -\sqrt{2}s\psi c\psi\\ 1 & s^2\psi & \sqrt{2}s\psi c\psi & c^2\psi \end{array}\,, \qquad (20)$$

where $c\psi=\cos\frac{1}{2}\beta$, $s\psi=\sin\frac{1}{2}\beta$. Results for the transformation elements $\langle M|m\rangle$ are obtained from Eq. (20) by transposing rows and columns. By considering the appropriate expansions it follows that

$$\langle M|=\sum_m\langle M|m\rangle\langle m|\,,|m\rangle=\sum_m\langle M|m\rangle|M\rangle\,,$$
$$\langle m|=\sum_M\langle m|M\rangle\langle M|\,. \qquad (21)$$

Expanding the state vector $|'\rangle$ in terms of the eigenstates $|M\rangle$ we have

$$|'\rangle=\sum_M a_M(t)|M\rangle+a_\mu(t)|\mu\rangle \qquad (22)$$

from which, using Eqs. (11) and (18), we find that

$$a_M=\sum_m a_m'\langle M|m\rangle\,, \quad \text{and} \quad a_m'=\sum_M\langle m|M\rangle a_M\,. \qquad (23)$$

The Schrödinger equation including the electric dipole perturbation may now be written as

$$i\hbar d|'\rangle/dt=(\mathcal{H}'+\mathcal{H}_e'')|'\rangle\,, \qquad (24)$$

where $\mathcal{H}_e''=U\mathcal{H}_e'U^\dagger$.

Hence substituting Eq. (22) into this equation and using the orthogonality and normalization of the states $|M\rangle$, we obtain the equations

$$i\hbar\dot{a}_M=(E_j+pM\hbar)a_M+\langle M|\mathcal{H}_e''|\mu\rangle a_\mu-\gamma_M/2a_M\,,$$
$$i\hbar\dot{a}_\mu=E_{j'}a_\mu+\sum_M\langle\mu|\mathcal{H}_e''|M\rangle a_M-\gamma_\mu/2a_\mu\,, \qquad (25)$$

where $E_j$ and $E_{j'}$ are the energies of the upper and lower levels for the degenerate case. Equation (25) may now be transformed to an interaction representation by putting

$$b_M(t)=a_M(t)e^{(i/\hbar)[E_j+pM\hbar]t} \qquad (26)$$

and similarly for $a_\mu$. Then substituting from Eqs. (18) and (21), the values of $|M\rangle$ and $\langle M|$ in terms of $|m\rangle$ and $\langle m|$, respectively, and using Eq. (14), we obtain the results

$$\dot{b}_M=(i\hbar)^{-1}\sum_m e^{im\omega_0 t}\langle M|m\rangle\langle m|\mathcal{H}_e|\mu\rangle\times e^{(i/\hbar)[E_j+pM\hbar-E_{j'}]t}b_\mu-\gamma_M/2b_M\,,$$
$$\dot{b}_\mu=(i\hbar)^{-1}\sum_{m,M}e^{-im\omega_0 t}\langle m|M\rangle\langle\mu|\mathcal{H}_e|m\rangle\times e^{-(i/\hbar)[E_j+pM\hbar-E_{j'}]t}b_M-\gamma_\mu/2b_\mu\,, \qquad (27)$$

in which $m$ and $M$ take values from $-J$ to $+J$, and where the relations between the probability amplitudes for the various representations used, which will determine the initial conditions, are given by Eqs. (12), (23), and (26).

Equation (27), together with these relations, represents the solution to the problem of simultaneous time-dependent electric and magnetic dipole perturbations on an atom. They are applicable to the case where the

[17] E. P. Wigner, *Group Theory and Its Application to the Quantum Mechanics of Atomic Spectra* (Academic Press Inc., New York, 1959), p. 167.

total angular momentum $j$ of the lower state is zero, but may be generalized immediately to that where the lower level has a finite $j$ value, provided the $g$ values of the upper and lower levels are the same. Such a restriction is necessary since the axis of quantization depends on the angle $\beta$ given by $\tan\beta=\gamma H_1/(\omega-\omega_0)$, where $\omega=geH/2mc$. The angle $\beta$ would thus be different for these levels if the $g$ values were different. In this event the approximation would be made that the effect of the rf field on one or other of the levels was negligible. Considering then equal $g$ values for the upper and lower levels we may write the generalized equations as

$$\begin{aligned}\dot{b}_M&=(i\hbar)^{-1}\textstyle\sum_{m\mu K} e^{i\omega_0 t(m-\mu)}b_K e^{(i/\hbar)[E_j+pM\hbar-E_{j'}-p\hbar K]t}\\&\quad\times\langle M|m\rangle\langle m|\mathcal{H}_e|\mu\rangle\langle\mu|K\rangle-\gamma_M/2b_M,\\\dot{b}_K&=(i\hbar)^{-1}\textstyle\sum_{m\mu M} e^{-i\omega_0 t(m-\mu)}b_M\\&\quad\times e^{-(i/\hbar)[E_j+pM\hbar-E_{j'}-p\hbar K]t}\langle K|\mu\rangle\langle\mu|\mathcal{H}_e|m\rangle\\&\quad\times\langle m|M\rangle-\gamma_K/2b_K,\end{aligned}\tag{28}$$

where

$$|K\rangle=\textstyle\sum_\mu\langle\mu|K\rangle|\mu\rangle,\ |\mu\rangle=\sum_K\langle K|\mu\rangle|K\rangle\tag{29}$$

and relations similar to those in Eqs. (12), (23), and (26) hold for $b_k(t)$. The coefficients $\langle\mu|K\rangle$, etc., are deduced from Eq. (19) for the given $j$ value. For a lower laser level with $j=2$, there would be $(2j+1)^2=25$ such coefficients.

To check the form of Eq. (27), assume first that there is no electric dipole perturbation, or $\mathcal{H}_e=0$. Then, from Eq. (27) we have

$$b_M(t)=b_M(O)e^{-\gamma_m/2t},\tag{30}$$

and using Eqs. (12), (23), and (26) we obtain

$$a_m e^{im\omega_0 t}=\textstyle\sum_M\langle m|M\rangle e^{-(i/\hbar)[E_j+pM\hbar-i\hbar\gamma_m/2]t}\times\sum_{m'}\langle M|m'\rangle a_{m'}(O).\tag{31}$$

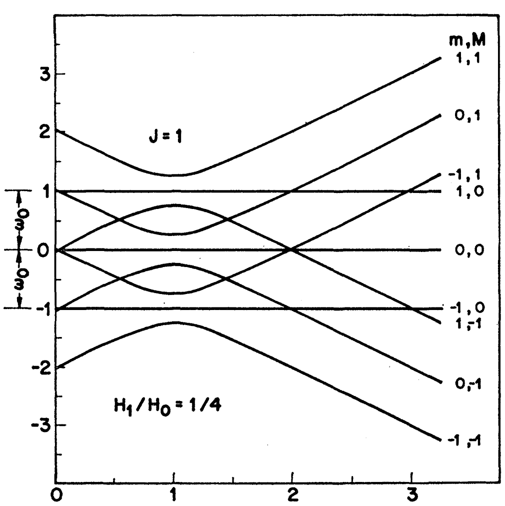


FIG. 3. Frequency diagram and level crossings for dc and rf magnetic fields acting on level $J=1$.

Supposing $a_{m'}(O)=\delta_{m'n}$; we obtain the result

$$a_m(t)=e^{-im\omega_0 t}\textstyle\sum_M\langle m|M\rangle\langle M|n\rangle\times e^{-(i/\hbar)[E_j+pM\hbar-i\hbar\gamma_m/2]t}.\tag{32}$$

Equation (32) gives the probability amplitude at time $t$ of the eigenstate $|m\rangle$, when the rf perturbation and damping terms have acted for a time $t$ on an atom which, at $t=0$, was in the eigenstate or Zeeman sublevel $|n\rangle$ of the excited state. It agrees with Eq. (17) of Ref. 7 and is analogous to the well-known Majorana formula[18] used for such considerations, within this case an extension to include the effects of the decay of the excited states. Secondly, suppose that there is no rf perturbation applied to the Zeeman sublevels of the excited state. Then we have $b=\gamma H_1=0$, $p=[(\omega-\omega_0)^2+b^2]^{1/2}=(\omega-\omega_0)$, $\beta=0$, $\langle m|M\rangle=\langle M|m\rangle=\delta_{mM}$, and $\sum_M\langle m|M\rangle=\sum_m\langle M|m\rangle=1$. Equation (27) then reduces to the form

$$\begin{aligned}\dot{b}_m&=(i\hbar)^{-1}\textstyle\sum_\mu\langle m|\mathcal{H}_e|\mu\rangle e^{i/\hbar[E_m-E_\mu]t}b_\mu-\gamma_m/2b_m,\\\dot{b}_\mu&=(i\hbar)^{-1}\textstyle\sum_m\langle\mu|\mathcal{H}_e|m\rangle e^{-i/\hbar[E_m-E_\mu]t}b_m-\gamma_\mu/2b_\mu,\end{aligned}\tag{33}$$

which correctly represent the equations for the probability amplitudes of the Zeeman sublevels under the effects of an electric dipole perturbation and damping terms in the interaction representation. These results essentially verify that the Eq. (27) represents the solution for the effect of both rf and optical perturbations on a single atom.

### 3. DISCUSSION

Equation (32) gives the probability amplitude $a_m(t)$ of the state $|m\rangle$ for an atom subjected to the simultaneous action of dc and rf magnetic fields, the atom being in the state $|n\rangle$ at $t=0$. A similar result may be obtained from Eq. (31) for any other initial state of the atom at $t=0$. By analogy with the expression $a_m(t)\exp(-iE_m t/\hbar)$, for the probability amplitude in a stationary field, we may draw effective frequency or term diagrams for the more general case of both rf and dc magnetic fields. Such a frequency diagram was first discussed by Pryce[2] for the case where $J=\frac{1}{2}$ and was further elaborated in the work of Dodd and Series. These results follow immediately from Eq. (31), which is here deduced with particular reference to stimulated emission processes as in the gas laser. The levels may be drawn from the terms in the exponent of Eq. (32), where the splitting occurs symmetrically about the level $E_j/\hbar$. We thus have the terms

$$m\omega_0+pM,\quad\text{where}\quad p=[(\omega-\omega_0)^2+(\gamma H_1)^2]^{1/2},\tag{34}$$

for the various levels with $m$, $M$ taking the values $-J$ to $+J$. A term diagram for the excited state $J=1$ is shown in Fig. 3, together with the values of $(m,M)$ for the various terms. These levels should be weighted by the term $\langle m|M\rangle\langle M|n\rangle$ for an atom initially in the state

[18] E. Majorana, Nuovo Cimento 9, 43 (1932).

$|n\rangle$, or by similar terms deduced from Eq. (31) for any other initial state of the atom. These terms govern the populations of the various levels and depend on the angle $\beta$ or on the levels of the dc and rf magnetic fields. Such terms, together with the damping term $\gamma_a$ must be considered in the analysis of a particular situation, but the simple term diagram is very effective in showing the values of magnetic fields at which coherence effects between the various levels will be most pronounced. We note that Fig. 3 is drawn for an assumed value of $H_1/H_0=\frac{1}{4}$; for lower values the shape of the curves would be affected most in the region $H/H_0=1$, but otherwise would not be changed unduly in other regions. $H_0$ is the value of dc magnetic field for which $\omega_0$, the rf frequency, is the Larmor frequency.

The quantum numbers $M$ correspond to the eigenstates in the rotating coordinate system in which the Hamiltonian is independent of time. Transitions between the levels due to the rf magnetic field are thus those in which $m$ changes but $M$ remains unchanged. Thus, supposing the atom is excited to the state (0,0) by suitable resonance radiation, the rf perturbation will then establish coherence between the levels (1,0), (0,0), and $(-1, 0)$, and in the case of fluorescent radiation this coherence will be evidenced by the appearance of beats or modulation effects at frequencies $\omega_0$ and $2\omega_0$ in the detected light. Other possibilities for coherence exist at values of dc magnetic fields at which levels cross as at $H=\frac{1}{2}H_0$, $H_0$, $2H_0$, and $3H_0$ and are discussed in Ref. 7. For example, at a magnetic field $H=3H_0$, the levels (1,0) and $(-1, 1)$ cross. Thus, if the incident polarization of the resonance radiation is orientated so as to excite both these states $|m=1\rangle$ and $|m=-1\rangle$ coherently, the rf perturbation will establish coherence between all levels with $M=1$ and $M=0$, and in the fluorescent radiation modulation effects or beat frequencies up to $4\omega_0$ will be possible for appropriate orientations of the observed polarization. Here such modulation effects arise from the interference between the spontaneous radiation from levels $mM=1$, 1; 0, 1; $-1$, 0; 1, 0; 0, 0; and $-1$, 0; these emit frequencies each differing by $\omega_0$ and which are rendered coherent by the exciting radiation and the rf magnetic field acting between the Zeeman levels. Such considerations readily follow from Eq. (32), which gives the probability amplitude $a_m(t)$ both in phase and amplitude.

For induced emission, as in the gas laser, differences will occur since only those transitions which correspond to laser emission, or to resonances of the laser cavity, will be effective. Thus, referring to Fig. 3, suppose the atom is in the state (0,0) and the laser frequency corresponds to the separation between this level and the lower level $J=0$ considered here. The rf perturbation will then establish coherence between the levels (1,0), (0,0), and $(-1, 0)$, but only the frequency corresponding to the level (0,0) will occur with the dc magnetic field transverse to the laser axis. Resonances of the rf frequency corresponding to $H=H_0$, or the Larmor frequency, will be manifested by changes in the intensity of the laser, particularly for low values of $H_1$, the amplitude of the rf magnetic field, since levels $(-1, 1)$ and $(-1, -1)$ will be close to level $(-1, 0)$ at this point and will be induced to emit. Resonance effects on the intensity of the laser emission may thus be expected, but no beats or modulation effects at $\omega_0$ or $2\omega_0$ will be present since the resonator is assumed not to respond to such frequencies. Modulation effects will occur if the frequency $\omega_0$ corresponds to the axial mode separation of the resonator, since in this case transitions from the levels (1,0) and $(-1, 0)$ will also occur. Other effects will occur where levels cross, such as when $H=\frac{1}{2}H_0$, etc., the laser resonator can then be tuned to the corresponding frequency and will then operate on both transitions from the levels (0,1) and $(-1, 1)$, assuming a suitable orientation of the dc magnetic field. Such states which cross are also excited coherently by the atomic or electronic collision processes involved, and will also be induced to emit coherently at the specific laser frequency. Intensity variations and polarization changes are to be expected in such regions of the frequency diagram, but no beat frequencies at $\omega_0$, etc., will appear unless the frequency $\omega_0$ again corresponds to frequency separation between axial modes of the resonator. These represent the general considerations involved in the interpretation of the frequency diagram in Fig. 3 for induced emission and in the use of Eqs. (27) and (28). Figure 4 shows a similar but more general term diagram deduced from

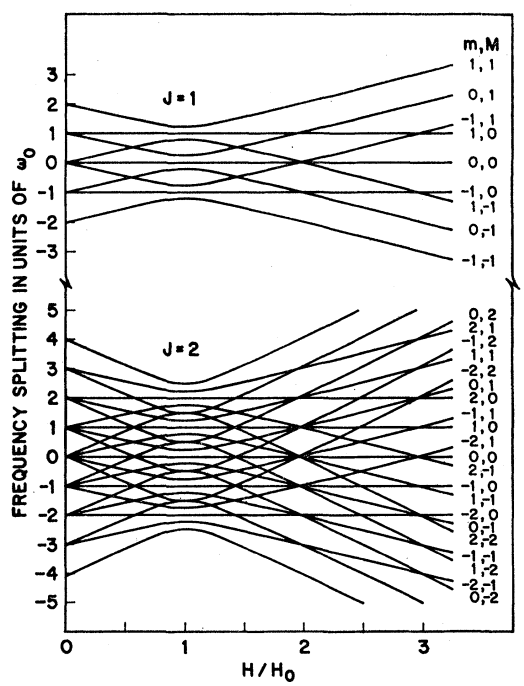


FIG. 4. Frequency diagram for action of dc and rf magnetic fields on the 1.153 $\mu$ He-Ne laser transition.

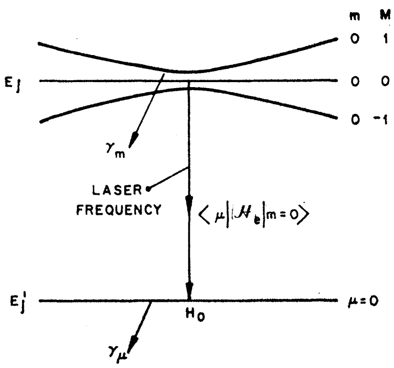


FIG. 5. Single laser transition $\langle \mu=0|\mathcal{H}_e|m=0\rangle$ and effective term diagram for $H\approx H_0$.

Eq. (28) for upper and lower levels with $J=1$ and $J=2$, respectively, where the $g$ values of the levels are assumed identical. This diagram is applicable to the 1.153 $\mu$ transition in the He-Ne laser and to other laser transitions. A few specific examples of the use of Eq. (27) will now be given. These will be restricted to a Zeeman splitting of only the upper level $J=1$, but similar considerations will apply to more complicated cases such as that shown in Fig. 4.

Consider first the case shown in Fig. 5 where the only laser transition or induced emission is that between the upper state $J=1$; $m$, $M=(0,0)$, and the lower state $J=0$. Let $\omega_{m\mu}=(E_j-E_{j'})/\hbar$, where here $m=\mu=0$; the matrix element involved for a transverse dc magnetic is then given by[19]

$$(\alpha jm|\mathbf{r}|\alpha' j+1m)=(\alpha j\vdots r\vdots\alpha' j+1)\mathbf{k}=C\mathbf{k}, \quad (34)$$

and the polarization will thus be linear and in the $z$ direction. Using Eq. (2) for the electric dipole perturbation together with the rotating wave approximation, Eq. (27) gives

$$\dot{b}_\mu=(eE/2i\hbar)\sum_M \langle 0|M\rangle e^{i(\Omega-pM)t}b_M-\gamma_\mu/2b_\mu, \quad (35)$$

where $\Omega=\omega_s-\omega_{m\mu}$, and we have omitted the constant $C$ in the electric dipole matrix element. There is a similar equation for the probability amplitudes of the upper levels from Eq. (27). The approximation is made that the decay of the $|M\rangle$ levels is primarily determined by the decay constant $\gamma_M$, which amounts to a small signal theory, the repopulation of the $M$ levels due to the laser emission is thus assumed small. Equation (27) then gives the result

$$b_M(t)=b_M(0)e^{-\gamma_m/2t}, \quad (36)$$

and substituting into Eq. (35) we obtain with $b_\mu(0)=0$ the result

$$b_\mu(t)=\frac{eE}{2i\hbar}e^{-\gamma_\mu/2t}\sum_M \frac{e^{i(\Omega-pM)t+\Gamma t}-1}{i(\Omega-pM)+\Gamma}\langle 0|M\rangle b_M(0), \quad (37)$$

[19] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, New York, 1935), p. 63.

where $\Gamma=\gamma_\mu/2-\gamma_m/2$, and the value of $b_M(0)$ must be determined by the initial condition from Eqs. (12), (23), and (26), which give

$$b_M(0)=a_M(0)=\sum_m a_m(0)\langle M|m\rangle. \quad (38)$$

For an atom in the state $|m=0\rangle$ at $t=0$, we thus obtain

$$b_M(0)=\langle M|0\rangle. \quad (39)$$

Substitution of the coefficients $\langle M|0\rangle$ and $\langle 0|M\rangle$, obtained from Eq. (20) and its transpose, into Eq. (37) gives the probability amplitude $b_\mu(t)$ for the atom in the state $|\mu=0\rangle$, and the emission of a photon with polarization in the direction $\mathbf{k}$.

Equation (37) gives the value of $b_\mu(t)$ for any value of $\Omega$, and for all values of $p$, which represents the effect of the rf magnetic field. Putting $\Omega=0$ and combining terms in Eq. (37) then gives

$$b_\mu(t)=\frac{eE}{2i\hbar}e^{-\gamma_\mu/2t}\left[\sin^2\beta\frac{\Gamma e^{\Gamma t}\cos pt+pe^{\Gamma t}\sin pt-\Gamma}{\Gamma^2+p^2}+\cos^2\beta\frac{e^{\Gamma t}-1}{\Gamma}\right] \quad (40)$$

and the total probability or steady condition for such a transition is then

$$P_s=\gamma_\mu\int_0^\infty |b_\mu(t)|^2dt. \quad (41)$$

Suppose that $\beta=0$, or no rf magnetic field is present, then Eqs. (40) and (41) give

$$P_s=(eE/\hbar)^2\gamma_\mu/[\gamma_m\gamma_\mu(\gamma_m+\gamma_\mu)], \quad (42)$$

which agrees with the general expression for such a transition probability in a two-level scheme,[20,21] under the small signal approximation. When the rf frequency $\omega_0$ equals the Larmor frequency $\omega=\gamma H$, $\beta=\pi/2$, and Eqs. (40) and (41) then give

$$P_s=\gamma_\mu\left(\frac{eE}{2\hbar}\right)^2\left[\frac{\gamma_m/2(\Gamma^2-p^2)+2\Gamma p^2}{\gamma_m^2+4p^2}+\frac{\Gamma^2+p^2}{2\gamma_m}+\frac{\Gamma^2}{\gamma_\mu}-\frac{2\Gamma(\Gamma\Gamma_1+p^2)}{\Gamma_1^2+p^2}\right]\Big/(\Gamma^2+p^2)^2, \quad (43)$$

which reduces to Eq. (42) for $p=0$, $\Gamma_1=\frac{1}{2}(\gamma_\mu+\gamma_m)$.

Similar results for general values of $\beta$ and $p$ may be deduced from Eq. (40). These special cases show that the probability $P_s$, which represents the intensity of the induced emission, will depend on the rf perturbation applied to the upper levels, and resonance effects will occur when $\omega_0$ corresponds to the Larmor frequency $\gamma H$. The polarization will correspond to the $\pi$ mode

[20] W. E. Lamb, Jr., and T. M. Sanders, Jr., Phys. Rev. **119**, 1901 (1960).
[21] W. R. Bennett, Jr., Phys. Rev. **126**, 580 (1962).

transition for the orientation considered here, and no beat frequencies will be observed. Other initial states of the atom at $t=0$ may be considered in a similar way using Eqs. (20) and (38). Thus, if the atom were initially in a coherent superposition of the states $|m\rangle$ with equal probabilities we would obtain the values

$$\begin{aligned} b_{-1}(0)&=[1+(\sin\beta)/\sqrt{2}]/\sqrt{3}\,,\\ b_0(0)&=(\cos\beta)/\sqrt{3}\,,\\ b_1(0)&=[1-(\sin\beta)/\sqrt{2}]/\sqrt{3}\,, \end{aligned} \tag{44}$$

and the analysis would then proceed by substituting these values into Eq. (37).

Consider now the situation shown in Fig. 6, where the frequency separation between the levels $(m,M)=(-1,0)$; $(1,0)$ equals the frequency interval between axial modes of the laser cavity. On tuning to these levels there will be two optical frequencies which will induce transitions between the upper and lower levels corresponding to $\Delta m=\pm1$ with matrix elements given by[19]

$$(\alpha jm|\mathbf{r}|\alpha' j+1m\pm1)=\mp C1/\sqrt{2}(\mathbf{i}\pm i\mathbf{j}) \tag{45}$$

for the laser axis along the $z$ direction. Thus, transitions $\Delta m=+1$ have an oscillating electric dipole $\mathbf{i}-i\mathbf{j}$ and give rise to right-hand circular polarization, while $\Delta m=-1$ corresponds to $\mathbf{i}+i\mathbf{j}$ and hence to left-hand circular polarization. For the transition $m=-1\rightarrow0$, Eq. (27) becomes

$$\dot{b}_\mu=(i\hbar)^{-1}\sum\nolimits_M\langle-1|M\rangle\langle0|\mathcal{H}_e|-1\rangle \times e^{-i/\hbar[W+pM\hbar-\omega_0\hbar]t}b_M-\gamma_\mu/2b_\mu\,, \tag{46}$$

where $W=E_j-E_{j'}$. Hence, with $\hbar\omega_{-1\mu}=W-\hbar\omega_0$ and $\Omega_1=\omega_s-\omega_{-1\mu}$, together with the small signal approximation, we obtain

$$b_\mu=\frac{eE}{2\sqrt{2}i\hbar}(\mathbf{i}-i\mathbf{j})e^{-\gamma_\mu/2t}\sum_M\frac{e^{i(\Omega_1-pM)t+\Gamma t}-1}{i(\Omega_1-pM)+\Gamma} \times\langle-1|M\rangle b_M(0)\,. \tag{47}$$

Similarly, we obtain for the transition $\Delta m=-1$ the result

$$b_\mu=-\frac{eE}{2\sqrt{2}i\hbar}(\mathbf{i}+i\mathbf{j})e^{-\gamma_\mu/2t}\sum_M\frac{e^{i(\Omega_2-pM)t+\Gamma t}-1}{i(\Omega_2-pM)+\Gamma} \times\langle1|M\rangle b_M(0)\,, \tag{48}$$

where $\Omega_2=\omega_{s'}-\hbar^{-1}(W+\hbar\omega_0)$, and hence $\omega_{s'}=\omega_s+2\omega_0$. Both $\Omega_1$ and $\Omega_2$ will be zero when the laser is tuned to the line centers. From Eqs. (47) and (48) we see that two oppositely circularly polarized transitions will occur, and that light beats at a frequency $2\omega_0$ will be obtained on detection. This corresponds to the modulation effects observed in the case of spontaneous emission as discussed earlier. From a consideration of Eqs. (47) and (48) we again see that the intensities of these transitions, and hence the beat amplitude, will vary with the parameter $p$ in a similar way to that discussed with reference to Eq. (37).

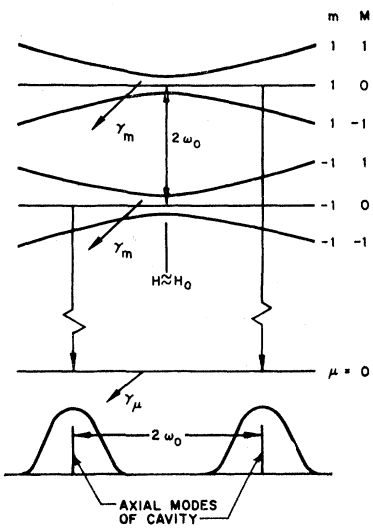


FIG. 6. Two laser transitions $\langle\mu|\mathcal{H}_e|m=1\rangle$ and $\langle\mu|\mathcal{H}_e|m=-1\rangle$ and effective term diagram for $H\approx H_0$.

As a final example we consider the point $H=2H_0$ in Fig. 3 at which the levels (0,0), $(-1,1)$, and $(1,-1)$ cross. Coherence is thus imparted to all levels $(m,M)$ in the diagram by the rf magnetic field. All transitions at the crossover point corresponding to $m=0,\pm1$ may be induced by a single laser frequency $\omega_s\approx W/\hbar$. Consider the laser oriented along the $z$ axis so that only the transitions $\Delta m=\pm1$ will be induced. At such a crossover point the atom will be excited by the collision process into a coherent mixture of the states (0,0), $(-1,1)$, and $(1,-1)$, so that the resulting probability amplitudes of the transitions $\Delta m=\pm1$ may be combined in a coherent way and hence, the probability and resultant polarization determined. The half-intensity widths of such transitions which crossover will be associated with the natural linewidths in contrast to other transitions which do not cross, where the linewidth will be determined by the magnitude of the rf field if this is large enough.

Equation (27) with both transitions $\Delta m=\pm1$ now becomes

$$\dot{b}_\mu=(eE/2i\hbar)\Big[\sum\nolimits_M\langle\mu|\mathcal{H}_e|-1\rangle\langle-1|M\rangle b_M \times e^{-i/\hbar(W+pM\hbar-\hbar\omega_0)t}+\sum\nolimits_M\langle\mu|\mathcal{H}_e|1\rangle\langle1|M\rangle b_M \times e^{-i/\hbar(W+pM\hbar+\hbar\omega_0)t}\Big]-\gamma_\mu/2b_\mu\,, \tag{49}$$

and the frequency $\omega_s$ of the inducing radiation is in the vicinity of $W/\hbar$. At the crossover point $H\approx2H_0$, and hence $p\approx\omega_0$ if $\gamma H_1$ is small. It is thus clear that the inducing laser radiation will have the greatest effect on the levels $(m,M)=(-1,1)$, $(1,-1)$, assuming only one effective cavity mode, since levels of the same $m$

but different $M$ are separated in frequency by $\omega_0$, which would be greater in general than the natural linewidths of the transitions. Only the values $M=1$ and $M=-1$ thus need to be considered in the first and second terms on the right-hand side of Eq. (49), respectively. Letting $\omega_s=W/\hbar$, and taking the rotating wave approximation in Eq. (2), we obtain

$$\dot{b}_\mu=(eE/2i\hbar)[\langle\mu|\mathbf{r}|-1\rangle\langle-1|M=1\rangle b_1 e^{i\Omega t}+\langle\mu|\mathbf{r}|1\rangle\langle 1|M=-1\rangle b_{-1}e^{-i\Omega t}]-\gamma_\mu/2t, \quad (50)$$

where $\Omega=\omega_0-p$. In the small signal approximation with $b_M$ given by Eq. (30), and with the matrix elements in Eq. (45), we find

$$b_\mu(t)=(eE/2\sqrt{2}\hbar)e^{-\gamma_\mu/2t}\left[-(\mathbf{i}-i\mathbf{j})\left(\frac{e^{(\Gamma+i\Omega)t}-1}{\Gamma+i\Omega}\right)\times\langle-1|M\rangle b_1(0)+(\mathbf{i}+i\mathbf{j})\left(\frac{e^{(\Gamma-i\Omega)t}-1}{\Gamma-i\Omega}\right)\times\langle 1|M\rangle b_{-1}(0)\right]. \quad (51)$$

Further reduction of Eq. (51) depends on the coefficients $\langle-1|M\rangle b_1(0)$ and $\langle 1|M\rangle b_{-1}(0)$, and hence on the initial state of the atom at $t=0$. For an atom in the state $|m=0\rangle$ at $t=0$, Eqs. (20) and (38) give the results

$$\langle m=-1|M=1\rangle b_1(0)=-\sqrt{2}\sin^3\beta/2\cos\beta/2,$$
$$\langle m=1|M=-1\rangle b_{-1}(0)=\sqrt{2}\sin^3\beta/2\cos\beta/2, \quad (52)$$

while if the initial state is a coherent superposition of the states $|m\rangle$ with equal probabilities, we obtain similarly

$$\langle m=-1|M=1\rangle b_1(0)=\sin^2\beta/2[1+(\sin\beta)/\sqrt{2}]/\sqrt{3}$$
$$\langle m=1|M=-1\rangle b_{-1}(0)=\sin^2\beta/2[1-(\sin\beta)/\sqrt{2}]/\sqrt{3}. \quad (53)$$

The initial probability amplitudes of the states $(-1, 1)$, and $(1, -1)$ thus depend on the angle $\beta$, and if this is small such levels will be weakly populated. Substituting Eqs. (52) and (53) into Eq. (51) shows that the resulting polarization will be linearly or elliptically polarized respectively, depending on the initial state of the atom. A rotation of the plane of polarization will also occur as $\Omega$ increases. This is related to the Hanle effect[22] at small dc magnetic fields where the states $|m\rangle$ overlap, and there is no rf field present.[23] At the cross-over point of the levels $(-1, 1)$ and $(1, -1)$, the dc magnetic field $H\approx 2H_0$, and $\omega=2\omega_0$, hence

$$\Omega=\omega_0-p\approx\gamma H_1^2/2H_0,$$
$$\tan\beta=H_1/H_0. \quad (54)$$

Hence, in general, depending on the lifetimes of the states, such effects will be small unless $H_1$ is comparable with $H_0$. This is analogous to the case of a strong dc field along the $z$ axis, even at the Larmor frequency resonance of the rf, and means that in this event the mixing of the states will be small. Figure 3 was drawn for $H_1/H_0$ equal to $\frac{1}{4}$, but smaller values of $H_1$ should give measurable effects in some instances. Other cross-over points in the region of $H/H_0=\frac{1}{2}, \frac{3}{2}, 3$ could be considered in a similar way by suitable orientation of the laser so that transitions $\Delta m=0, \pm 1$ are possible.

Similar results may be obtained for the more general level scheme shown in Fig. 4. Here, since the lower levels are also split to a similar degree, more transitions at a given frequency $\omega_s$ can be induced between the upper and lower states, but the results would be analogous to those already derived. These will be considered at some future time in conjunction with experimental results. It also follows that Eqs. (27) and (28) are applicable for absorption processes as well as emission. In this event we would determine the probability amplitudes $b_M(t)$ for particular initial values of $b_\mu(0)$ or $b_K(0)$. Finally, the effects of both rf magnetic fields and optical perturbations on a single atom have been considered in relation to intensity and polarization changes in the radiation. In the actual gas laser there are great numbers of atoms with a velocity distribution corresponding to the temperature of the emitting atoms. However, any treatment of the macroscopic system under such conditions must reflect the characteristics of the microscopic systems which comprise the ensemble. Various initial states of the atom have been assumed in the theory, and such double-resonance effects represents a potential method for the investigation of the excitation and decay processes involved in the laser.

[22] W. Hanle, Z. Physik **30**, 93 (1924).
[23] W. Culshaw and J. Kannelaud, Bull. Am. Phys. Soc. **9**, 65 (1964).