any time. This type of modulation can be neglected when small input signals are considered and is not important under most circumstances. This shot effect and also the effect of power flow through the cavity on the frequency dependence of the amplification will be discussed in more detail in a subsequent paper.

Amplification may also be accomplished using one wave guide as both input and output, and the noise figure of such an amplifier can also approach unity. The amplified output signal might be coupled out and detected through a directional coupler, which would have to have a fairly small coupling so that little of the input power was lost to it. Then so long as the amplified input noise appearing at the detector was large compared to kT, the noise figure of this amplifier would be small.

The maser amplifier may be useful in a restricted range of applications in spite of its narrow band width because of its potentially low noise figure. For example, suppose that the signal to be amplified came from outer space, where the temperature is only a few degrees absolute. Then by making the coupling through the cavity fairly large so that little noise is contributed by the cavity itself, amplification should be attainable while keeping the noise figure, *based on the temperature of the signal source*, fairly low. This might prove to have a considerable advantage over electronic amplifiers. It might also be possible to tune the frequency of a maser amplifier through the use of the Stark or Zeeman effects on the molecular transition frequencies.

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Resonance Transitions in Molecular Beam Experiments. I. General Theory of Transitions in a Rotating Magnetic Field*

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The time-dependent Schrödinger equation is solved formally for an atomic or molecular system which is subjected simultaneously to a rotating magnetic field of constant amplitude and angular velocity and to a constant magnetic field along the axis of rotation. The method yields the transition probabilities in terms of the solutions to an eigenvalue problem. This eigenvalue problem is solved both for (*a*) a normal Zeeman effect and for (*b*) the case where a transition from a given level is isolated in frequency from other transitions from the same level. Case (*a*) is exactly soluble and yields a solution which is shown to be the same as that of Bloch and Rabi, but is in a form which is more convenient for integration over the velocity distribution. Case (*b*) must be solved by an approximate method which results in a prediction of multiple quantum transitions as observed by Kusch.

IN the C-field of a molecular beams apparatus, a system (e.g., an atom or molecule) is subjected, in a region of constant magnetic field, to an oscillating magnetic field. The experimenter observes, as a function of frequency, a quantity proportional to the number of atoms which have left their initial states.

We will be dealing here with the case in which the constant and oscillating fields are perpendicular to each other. In this case, the solution is greatly simplified[1] by replacing the oscillating field by one which is rotating about the direction of the constant field. One must then correct for the effect of the other rotating component,[2] but the correction is usually small. In the present paper, we shall also explicitly assume that the constant magnetic field is uniform and that the rotating field is uniform in both amplitude and phase.

With these assumptions, one may, for a normal Zeeman effect, solve exactly for the probability of a transition being induced by the rotating field.[3] In other cases, however, some approximate method must be used. Allowed transitions may usually be treated by assuming that only the two levels involved interact,[4] but in the case of multiple quantum transitions which take place only by virtue of the existence of intermediate states, it is necessary to have a more general approach.

The approach we shall develop here has the virtue of not only yielding expressions for the probabilities of multiple quantum transitions but also giving corrections to the single quantum transition probabilities and a convenient expression for the exact solution of the normal Zeeman effect problem.

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[1] See, for example, I. I. Rabi, Phys. Rev. **51**, 652 (1937).
[2] F. Bloch and A. Siegert, Phys. Rev. **57**, 522 (1940).
[3] F. Bloch and I. I. Rabi, Revs. Modern Phys. **17**, 237 (1945); E. Majorana, Nuovo cimento **9**, 43 (1932).
[4] H. C. Torrey, Phys. Rev. **59**, 293 (1941).

1. FORMAL SOLUTION

The problem may be stated as follows: A system whose unperturbed Hamiltonian is invariant under spatial rotations enters a region in which there is a constant field of magnitude H_z in the z-direction and a rotating field of amplitude H_0 in the xy-plane. If the initial state of the system is $|(t=0)\rangle$, what is the probability that at a time $t=\tau$ it will be observed to have left that state?

We now proceed to solve this problem.

A. System in a Constant Magnetic Field

Let $\mathcal{H}_0$ be the unperturbed Hamiltonian, $\mathbf{J}$ the total angular momentum, $\mathbf{J}_1$, $\mathbf{J}_2$, $\cdots\mathbf{J}_h$ the individual spin or orbital angular momenta which make up $\mathbf{J}$, and g_1, g_2, $\cdots g_h$ the corresponding gyromagnetic ratios. Since $\mathcal{H}_0$ is invariant under spatial rotations it will commute with both J^2 and J_z. Its eigenstates may then be taken to be eigenstates of J^2, J_z, and of a set of commuting variable, Γ, which commute with J^2 and J_z. The eigenvalues will be independent of J_z. The eigenstates will then satisfy the equation

$$\mathcal{H}_0|\gamma,j,m\rangle=W_0(\gamma,j)|\gamma,j,m\rangle, \tag{1}$$

where γ, $\hbar j(j+1)$, and $m\hbar$ are the eigenvalues of Γ, J^2, and J_z, respectively.

If we consider the effect of the constant field alone, the perturbed Hamiltonian will be

$$\mathcal{H}_1=\mathcal{H}_0+H_z(\mu_0/\hbar)[g_1J_{1z}+g_2J_{2z}+\cdots+g_hJ_{hz}]. \tag{2}$$

Although $\mathcal{H}_1$ in general no longer commutes with Γ or J^2 it does still commute with J_z. The steady states in this field will then be states for which m is a good quantum number but γ and j are not. For each state $|\gamma,j,m\rangle$ there will be one steady state which approaches it as H_z approaches zero. We will use the notation $|(\gamma,j),m\rangle$ for this state. The state $|(\gamma,j)m\rangle$ will then satisfy the equation

$$\mathcal{H}_1|(\gamma,j),m\rangle=W_1(\gamma,j,m)|(\gamma,j),m\rangle, \tag{3}$$

with

$$|(\gamma,j),m\rangle\rightarrow|\gamma,j,m\rangle \quad \text{as} \quad H_z\rightarrow 0, \tag{4}$$

where $W_1(\gamma,j,m)$ is the perturbed energy.

B. System with Rotating Field Included. Elimination of the Time from the Hamiltonian

Now we must introduce the rotating magnetic field. If the x-axis is chosen in the direction of the field at $t=0$ we have

$$H_x=H_0\cos\omega t, \quad H_y=H_0\sin\omega t. \tag{5}$$

Then the total Hamiltonian is

$$\begin{aligned}\mathcal{H}&=\mathcal{H}_1+H_x(\mu_0/\hbar)[g_1J_{1x}+g_2J_{2x}+\cdots+g_hJ_{hx}]\\&\quad+H_y(\mu_0/\hbar)[g_1J_{1y}+g_2J_{2y}+\cdots+g_hJ_{hy}]\\&=\mathcal{H}_1+\tfrac{1}{2}(H_x+iH_y)(\mu_1/\hbar)[g_1(J_{1x}-iJ_{1y})+\cdots\\&\quad+g_h(J_{hx}-iJ_{hy})]+\tfrac{1}{2}(H_x-iH_y)(\mu_0/\hbar)\\&\quad\times[g_1(J_{1x}+iJ_{1y})+\cdots+g_h(J_{hx}+iJ_{hy})],\\&=\mathcal{H}_1+H_0(\mu_0/2\hbar)\{e^{i\omega t}[g_1(J_{1x}-iJ_{1y})+\cdots\\&\quad+g_h(J_{hx}-iJ_{hy})]+e^{-i\omega t}[g_1(J_{1x}+iJ_{1y})+\cdots\\&\quad+g_h(J_{hx}+iJ_{hy})]\}.\end{aligned} \tag{6}$$

Making use of the fact[5] that $\langle\gamma,j,m|J_{kx}\pm iJ_{ky}|\gamma',j'm'\rangle$ is different from zero only when m' is equal to $m\mp1$, we find that

$$\begin{aligned}&e^{\mp i\omega t}\langle\gamma,j,m|J_{kx}\pm iJ_{ky}|\gamma',j',m'\rangle\\&=e^{-im\omega t}\langle\gamma,j,m|J_{kx}\pm iJ_{ky}|\gamma',j',m'\rangle e^{im'\omega t}\\&=\langle\gamma,j,m|e^{-(i/\hbar)J_z\omega t}(J_{kx}\pm iJ_{ky})e^{(i/\hbar)J_z\omega t}|\gamma',j',m'\rangle,\end{aligned} \tag{7}$$

and, therefore

$$e^{\mp i\omega t}(J_{kx}\pm iJ_{ky})=e^{-(i/\hbar)J_z\omega t}(J_{kx}\pm iJ_{ky})e^{(i/\hbar)J_z\omega t}. \tag{8}$$

We thus obtain the expression

$$\begin{aligned}\mathcal{H}&=\mathcal{H}_1+H_0(\mu_0/2\hbar)e^{-(i/\hbar)J_z\omega t}\{[g_1(J_{1x}-iJ_{1y})+\cdots\\&\quad+g_h(J_{hx}-iJ_{hy})]+[g_1(J_{1x}+iJ_{1y})+\cdots\\&\quad+g_h(J_{hx}+iJ_{hy})]\}e^{(i/\hbar)J_z\omega t},\\&=e^{-(i/\hbar)J_z\omega t}\{\mathcal{H}_1+H_0(\mu_0/\hbar)\\&\quad\times[g_1J_{1x}+\cdots+g_hJ_{hx}]\}e^{(i/\hbar)J_z\omega t}\end{aligned} \tag{9}$$

since $\mathcal{H}_1$ commutes with J_z.

We may make use of the fact that the expression in the brackets does not contain the time to reduce the solution of our problem to the solution of a problem whose Hamiltonian is independent of the time.

If $|\ \rangle$ is a state of the system it will satisfy the time-dependent Schrödinger equation:

$$i\hbar(d/dt)|\ \rangle=\mathcal{H}|\ \rangle. \tag{10}$$

Then if we make the substitution

$$|'\rangle=e^{(i/\hbar)J_z\omega t}|\ \rangle, \tag{11}$$

$|'\rangle$ will satisfy the equation

$$\begin{aligned}&i\hbar(d/dt)|'\rangle\\&=e^{(i/\hbar)J_z\omega t}\{i\hbar(d/dt)|\ \rangle\}-\omega J_ze^{(i/\hbar)J_z\omega t}|\ \rangle\\&=e^{(i/\hbar)J_z\omega t}\mathcal{H}|\ \rangle-\omega J_ze^{(i/\hbar)J_z\omega t}|\ \rangle\\&=\{(\mathcal{H}_1-\omega J_z)+(\mu_0H_0/\hbar)\\&\quad\times[g_1J_{1x}+g_2J_{2x}+\cdots+g_hJ_{hx}]\}|'\rangle\\&=\mathcal{H}'|'\rangle,\end{aligned} \tag{12}$$

[5] See, for example, E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, London, 1951), Eq. 9311.

where

$$\mathcal{H}'=(\mathcal{H}_1-\omega J_z)+(\mu_0 H_0/\hbar)[g_1 J_{1x}+\cdots+g_h J_{hx}] \quad (13)$$

is independent of the time.

C. Solution of the Equations of Motion. Calculation of Transition Probabilities[6]

Just as in any such problem, we may find the eigenstates of the operator by solving the secular equation:

$$\det(\mathcal{H}'-\lambda I)=0 \quad (14)$$

for the eigenvalues λ and then solving a set of linear equations for the corresponding state vectors $|\lambda\rangle$. In general, there will be one eigenvalue $\lambda_{\gamma jm}$ for each state $|\gamma,j,m\rangle$ or $|(\gamma,j),m\rangle$.

Now given any state $|\ \rangle$ at $t=0$, we may expand $|'\rangle$ in terms of the states $|\lambda\rangle$:

$$|'\rangle=\sum_\lambda|\lambda\rangle\langle\lambda|'\rangle, \quad (15)$$

where the coefficients $\langle\lambda|'\rangle$ will be functions of the time. Then the equation of motion

$$i\hbar(d/dt)|'\rangle=\mathcal{H}'|'\rangle, \quad (12)$$

becomes

$$\sum_\lambda|\lambda\rangle\{i\hbar(d/dt)\langle\lambda|'\rangle\}=\sum_\lambda \mathcal{H}'|\lambda\rangle\langle\lambda|'\rangle$$
$$=\sum_\lambda|\lambda\rangle\lambda\langle\lambda|'\rangle, \quad (16)$$

or

$$i\hbar(d/dt)\langle\lambda|'\rangle=\lambda\langle\lambda|'\rangle, \quad (17)$$

with the solution

$$\langle\lambda|'\rangle=e^{-(i/\hbar)\lambda t}\langle\lambda|'(t=0)\rangle=e^{-(i/\hbar)\lambda t}\langle\lambda|(t=0)\rangle. \quad (18)$$

We finally obtain

$$|'\rangle=\sum_\lambda|\lambda\rangle e^{-(i/\hbar)\lambda t}\langle\lambda|(t=0)\rangle \quad (19)$$

or

$$|\ \rangle=e^{-(i/\hbar)J_z\omega t}\sum_\lambda|\lambda\rangle e^{-(i/\hbar)\lambda t}\langle\lambda|(t=0)\rangle. \quad (20)$$

We must now calculate the probability that a system initially in a state $|(t=0)\rangle$ has left that state by the time τ. We may write this as

$$P[|(t=0)\rangle\rightarrow]=1-|\langle(t=0)|(t=\tau)\rangle|^2, \quad (21)$$

where, by (20),

$$\langle(t=0)|(t=\tau)\rangle$$
$$=\sum_\lambda\langle(t=0)|e^{-(i/\hbar)J_z\omega\tau}|\lambda\rangle e^{-(i/\hbar)\lambda\tau}\langle\lambda|(t=0)\rangle. \quad (22)$$

One ordinarily deals with initial states for which J_z is quantized. In this case, the initial state satisfies an equation of the form

$$J_z|(t=0)\rangle=\mu\hbar|(t=0)\rangle, \quad (23)$$

so that

$$\langle(t=0)|(t=\tau)\rangle=e^{-i\mu\omega\tau}\sum_\lambda|\langle(t=0)|\lambda\rangle|^2 e^{-(i/\hbar)\lambda\tau}. \quad (24)$$

Then

$$P[|(t=0)\rangle\rightarrow]$$
$$=1-\sum_{\lambda,\lambda'}e^{(i/\hbar)(\lambda-\lambda')\tau}\times|\langle(t=0)|\lambda\rangle|^2|\langle(t=\mathrm{p})|\lambda'\rangle|^2$$
$$=1-\sum_{\lambda,\lambda'}\cos[(1/\hbar)(\lambda-\lambda')\tau]\times|\langle(t=0)|\lambda\rangle|^2|\langle(t=0)|\lambda'\rangle|^2$$
$$=1-\sum_\lambda|\langle(t=0)|\lambda\rangle|^2\sum_{\lambda'}|\langle(t=0)|\lambda'\rangle|^2$$
$$+\sum_{\lambda,\lambda'}\{1-\cos[(1/\hbar)(\lambda-\lambda')\tau]\}\times|\langle(t=0)|\lambda\rangle|^2|\langle(t=0)|\lambda'\rangle|^2$$
$$=4\sum_{\lambda>\lambda'}|\langle(t=0)|\lambda\rangle|^2|\langle(t=0)|\lambda'\rangle|^2\times\sin^2[(1/2\hbar)(\lambda-\lambda')\tau]. \quad (25)$$

D. Physical Interpretation of the Result

The above discussion may be interpreted in the following way.

The substitution (11) was a transformation from a fixed frame of reference to one which is rotating with angular velocity ω about the z-direction. In this frame of reference, the effective Hamiltonian is $\mathcal{H}'$ which is independent of the time. The steady states, $|\lambda\rangle$, of $\mathcal{H}'$ are thus the steady states of the system.

If the initial state is one of the eigenstates, the system will remain in this eigenstate and the transition probability will be zero. If, on the other hand, the eigenstate is a linear combination of two or more of the eigenstates, the final state will contain the same amount of each eigenstate, but the relative phases will be changed. This change in the phases will cause transitions to other linear combinations of the same eigenstates.

In the expression (25) for the transition probability, each term is the contribution due to the phase difference of a single pair of states, $|\lambda\rangle$ and $|\lambda'\rangle$. The effect of this phase difference is given by

$$4\sin^2[(1/2\hbar)(\lambda-\lambda')\tau]=|e^{-(i/\hbar)\lambda\tau}-e^{-(i/\hbar)\lambda'\tau}|^2. \quad (26)$$

The coefficient

$$|\langle(t=0)|\lambda\rangle|^2|\langle(t=0)|\lambda'\rangle|^2$$
$$=(\text{initial probability of eigenstate }|\lambda\rangle)\times(\text{initial probability of eigenstate }|\lambda'\rangle) \quad (27)$$

expresses the fact that the phase difference between these states can cause transitions only to the extent that *both* states are present initially.

[6] *Note added in proof.*—Mr. M. Hack has pointed out to me that Besset *et al.* [J. phys. radium **15**, 251 (1954)] similarly approach the problem of "multiple-flop" transitions in terms of the steady states of $\mathcal{H}'$. Mr. Hack has also independently applied this "spectral representation" to the problem of multiple quantum transitions (unpublished).

E. The Case of $|(t=0)\rangle = |(\gamma,j),m\rangle$. Selection Rule for Transitions

In the usual experiment, the system enters the radio-frequency field after it has already been in the constant magnetic field, so that our initial state should be taken to be one of the states $|(\gamma,j),m\rangle$. For this case, the formula for the transition probability becomes

$$P[|(\gamma,j),m\rangle\rightarrow]=4\sum_{\lambda<\lambda'}|\langle(\gamma,j),m|\lambda\rangle|^2 \times|\langle(\gamma,j),m|\lambda'\rangle|^2\sin^2[(1/2\hbar)(\lambda-\lambda')\tau]. \quad (28)$$

In Secs. 2 and 3, we shall derive the transition probabilities in the two cases which cover most experimental applications. It is nevertheless instructive to determine here the resonant frequencies which may generally be expected.

Unless an exact solution is possible, it is simplest to work in the representation in which $\mathcal{H}_1$ is diagonal. In this representation, we expand in terms of the $|(\gamma,j),m\rangle$, which also happen to be our initial states. According to the foregoing derivation, our effective Hamiltonian is

$$\mathcal{H}'=(\mathcal{H}_1-\omega J_z)+(\mu_0H_0/\hbar)[g_1J_{1x}+\cdots+g_hJ_{hx}], \quad (13)$$

with

$$\mathcal{H}_1=\mathcal{H}_0+(\mu_0H_z/\hbar)[g_1J_{1z}+\cdots+g_hJ_{hz}]. \quad (2)$$

The first term, $\mathcal{H}_1-\omega J_z$, of $\mathcal{H}'$ is completely diagonal in our chosen representation, while the remaining term is completely off-diagonal.

Under ordinary experimental conditions the amplitude of the oscillating field is much smaller than that of the constant field, so it is reasonable to treat the off-diagonal terms of $\mathcal{H}'$ (proportional to the rotating field amplitude H_0) as small perturbations. Thus, except when two or more levels are degenerate or nearly degenerate, the states $|(\gamma,j),m\rangle$ are good approximations to the eigenstates of $\mathcal{H}'$ and no transitions are to be expected.

We should now note that the diagonal elements

$$\langle(\gamma,j),m|\,\mathcal{H}'\,|(\gamma,j),m\rangle=W_1(\gamma,j,m)-m\hbar\omega \quad (29)$$

are linear functions of the angular frequency ω. The frequency may thus be chosen so as to produce degeneracy. Those frequencies at which degeneracy occurs will (for $H_0\ll H_z$) be the resonant frequencies. Our condition for degeneracy between the levels $|(\gamma,j),m\rangle$ and $|(\gamma',j'),m'\rangle$ is thus

$$\langle(\gamma,j),m|\,\mathcal{H}'\,|(\gamma,j),m\rangle=\langle(\gamma',j'),m'|\,\mathcal{H}'\,|(\gamma',j'),m'\rangle \quad (30)$$

or

$$(m-m')\hbar\omega=W_1(\gamma,j,m)-W_1(\gamma',j',m'). \quad (31)$$

At the rotational frequency given by Eq. (31), a resonance should occur involving transitions between the states $|(\gamma,j),m\rangle$ and $|(\gamma',j'),m'\rangle$.

Up till now we have spoken in terms of a rotating field, whose angular frequency can have either sign. For the actual oscillating field, the frequencies are given by[7]

$$|m-m'|\hbar\omega=|W_1(\gamma,j,m)-W_1(\gamma',j',m')|. \quad (32)$$

Resonances will be observed at the frequencies given by Eq. (32) only if the amplitude of the oscillating field is large enough to produce transitions in the available time.

2. APPLICATION TO NORMAL ZEEMAN EFFECT

A. Solution for Transition Probabilities

A normal Zeeman effect occurs when we may take $\mathcal{H}_1$ and $\mathcal{H}$ to be given by[8]

$$\mathcal{H}_1=\mathcal{H}_0+(g\mu_0H_z/\hbar)J_z,$$
$$\mathcal{H}=\mathcal{H}_1+(g\mu_0/\hbar)\{H_xJ_x+H_yJ_y\}. \quad (33)$$

In this case, we have

$$|(\gamma,j),m\rangle=|\gamma,j,m\rangle,$$
$$\mathcal{H}_1|\gamma,j,m\rangle=(E_0+mg\mu_0H_z)|\gamma,j,m\rangle, \quad (34)$$

and

$$\mathcal{H}'=\mathcal{H}_0+[(g\mu_0H_z/\hbar)-\omega]J_z+(g\mu_0H_0/\hbar)J_x. \quad (35)$$

Let us introduce the following notation:

$$a=g\mu_0H_z/\hbar,\quad b=g\mu_0H_0/\hbar,$$
$$\cos\varphi=(a-\omega)/[(a-\omega)^2+b^2]^{\frac{1}{2}},$$
$$\sin\varphi=b/[(a-\omega)^2+b^2]^{\frac{1}{2}}, \quad (36)$$
$$\theta=[(a-\omega)^2+b^2]^{\frac{1}{2}}\tau,$$

where τ is the time the system is in the oscillating field. Then

$$\mathcal{H}'=\mathcal{H}_0+(a-\omega)J_z+bJ_x$$
$$=\mathcal{H}_0+[(a-\omega)^2+b^2]^{\frac{1}{2}}[\cos\varphi J_z+\sin\varphi J_x] \quad (37)$$
$$=\mathcal{H}_0+[(a-\omega)^2+b^2]^{\frac{1}{2}}J_{z'},$$

where the z'-axis is as shown in Fig. 1. The eigenvalues of $J_{z'}$ are $\mu\hbar$ ($\mu=-j,\ -j+1,\ \cdots,\ j$). Then the eigenvalues and eigenvectors of $\mathcal{H}'$ will be

$$\lambda_{\gamma j\mu}=E_0(\gamma,j)+[(a-\omega)^2+b^2]^{\frac{1}{2}}\mu\hbar,$$
$$|\lambda_{\gamma j\mu}\rangle=|\gamma,j,J_{z'}=\mu\hbar\rangle, \quad (38)$$

[7] We should note there that, even though Eq. (31) includes all resonances due to a rotating field, Eq. (32) does not include all possible resonances, due to an oscillating field. An example of a resonance which is not predicted by (32) is that due to the two-quantum transition $a\leftrightarrow i\leftrightarrow b$ where $m_a=m_b$, $m_i=m_a\pm1$. In this case, the states $|a\rangle$ and $|i\rangle$ are connected by one rotating component, while the states $|i\rangle$ and $|b\rangle$ are connected by the opposite component. As a result there should be a resonance at the frequency $\hbar\omega=|(W_a-W_b)/2|$. This resonance will have a large peak intensity when $W_i\sim\frac{1}{2}(W_a+W_b)$.

[8] This may be true for several possible reasons: (1) All the g_k's may be equal. (2) Some of the J_k's may be identically equal to zero for the states under consideration and the remaining g_k's may be equal. (3) The magnetic field may be too weak to decouple certain of the angular momenta from each other with the result that Eq. (33) turns out to be a good approximation. Except in the first case, we should then take $g=g(\gamma,j)$.

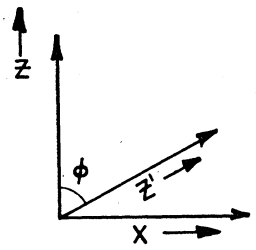

FIG. 1. Relation between the z- and z'-axes.

where the states $|\gamma,j,\mu\rangle$ are quantized along the z'-direction rather than the z-direction. The states $|\gamma,j,\mu\rangle$ may be obtained from the states $|\gamma,j,m\rangle$ by a rotation of the coordinate system through an angle φ. This rotation is effected by the irreducible representation of the rotation group corresponding to angular momentum j. In what follows, we shall use the notation of Wigner[9] for these operators.

The eigenfunctions of $\mathcal{H}'$ are given by

$$|\gamma,j,J_{z'}=\mu\hbar\rangle=\sum_m \mathfrak{D}^{(j)}(0,\varphi,0)_{\mu m}|\gamma,j,J_z=m\hbar\rangle. \quad (39)$$

We then have

$$\langle\gamma,\ j,J_z=m\hbar|\gamma,\ j,J_{z'}=\mu\hbar\rangle=\mathfrak{D}^{(j)}(0,\varphi,0)_{\mu m},$$
$$\langle\gamma,\ j,J_{z'}=\mu\hbar|\gamma,\ j,J_z=m\hbar\rangle=\mathfrak{D}^{(j)}(0,-\varphi,0)_{m\mu}. \quad (40)$$

The transition probabilities may now be found immediately:

$$P[|\gamma,j,m\rangle\rightarrow]=4\sum_{\mu>\mu'}|\mathfrak{D}^{(j)}(0,\varphi,0)_{\mu m}|^2 \times|\mathfrak{D}^{(j)}(0,\varphi,0)_{\mu' m}|^2\sin^2\tfrac{1}{2}(\mu-\mu')\theta. \quad (41)$$

B. Proof of Equivalence with the Result of Bloch and Rabi

In the paper of Bloch and Rabi,[10] the probability of a transition from $|\gamma,j,m\rangle$ to $|\gamma,j,m'\rangle$ is given by $|T_{mm'}|^2$, where (see Appendix of this paper)

$$T_{mm'}=T_{m'm}=\mathfrak{D}^{(j)}(R)_{mm'},$$
$$\mathfrak{D}^{(\frac{1}{2})}(R)=\begin{pmatrix}\cos(\theta/2)+i\cos\varphi\sin(\theta/2) & i\sin\varphi\sin(\theta/2)\\ i\sin\varphi\sin(\theta/2) & \cos(\theta/2)-i\cos\varphi\sin(\theta/2)\end{pmatrix}. \quad (42)$$

The same probability is given in the present theory by $|\langle\gamma,j,m'|(t=\tau)\rangle|^2$, where $|(t=0)\rangle=|\gamma,j,m\rangle$. Using Eq. (20), we get [see Eq. (113)]

$$\begin{aligned}\langle\gamma,j,m'|(t=\tau)\rangle&=\sum_\mu\langle\gamma,j,m'|\exp[-(i/\hbar)J_z\omega\tau]|\gamma,j,\mu\rangle\exp[-(i/\hbar)\{E_0+\mu\hbar[(a-\omega)^2+b^2]^{\frac{1}{2}}\}\tau]\langle\gamma,j,\mu|\gamma,j,m\rangle\\ &=\exp\{-(i/\hbar)[E_0+m'\hbar\omega]\tau\}\sum_\mu\mathfrak{D}^{(j)}(0,\varphi,0)_{\mu m'}e^{-i\mu\theta}\mathfrak{D}^{(j)}(0,-\varphi,0)_{m\mu}\\ &=\exp\{-(i/\hbar)[E_0+m'\hbar\omega]\tau\}\sum_\mu\mathfrak{D}^{(j)}(0,\varphi,0)_{\mu m'}\mathfrak{D}^{(j)}(0,-\varphi,-\theta)_{m\mu}\\ &=\exp\{-(i/\hbar)[E_0+m'\hbar\omega]\tau\}\mathfrak{D}^{(j)}(S)_{mm'},\end{aligned} \quad (43)$$

where

$$\begin{aligned}\mathfrak{D}^{(\frac{1}{2})}(S)&=\mathfrak{D}^{(\frac{1}{2})}(0,-\varphi,-\theta)\mathfrak{D}^{(\frac{1}{2})}(0,\varphi,0)\\ &=\begin{pmatrix}\cos(\varphi/2)e^{-i\theta/2} & -\sin(\varphi/2)e^{i\theta/2}\\ \sin(\varphi/2)e^{-i\theta/2} & \cos(\varphi/2)e^{i\theta/2}\end{pmatrix}\begin{pmatrix}\cos(\varphi/2) & \sin(\varphi/2)\\ -\sin(\varphi/2) & \cos(\varphi/2)\end{pmatrix}\\ &=\begin{pmatrix}\sin^2(\varphi/2)e^{i\theta/2}+\cos^2(\varphi/2)e^{-i\theta/2} & -\sin(\varphi/2)\cos(\varphi/2)(e^{i\theta/2}-e^{-i\theta/2})\\ -\sin(\varphi/2)\cos(\varphi/2)(e^{i\theta/2}-e^{-i\theta/2}) & \cos^2(\varphi/2)e^{i\theta/2}+\sin^2(\varphi/2)e^{-i\theta/2}\end{pmatrix}\\ &=\begin{pmatrix}\cos(\theta/2)-i\cos\varphi\sin(\theta/2) & -i\sin\varphi\sin(\theta/2)\\ -i\sin\varphi\sin(\theta/2) & \cos(\theta/2)+i\cos\varphi\sin(\theta/2)\end{pmatrix}\\ &=[\mathfrak{D}^{(\frac{1}{2})}(R)]^{-1}=\mathfrak{D}^{(\frac{1}{2})}(R^{-1}).\end{aligned} \quad (44)$$

Equation (42) was used to obtain the last line.

Then $\mathbf{S}=\mathbf{R}^{-1}$ and

$$|\langle\gamma,\ j,\ m|(t=\tau)\rangle|^2=|\mathfrak{D}^{(j)}(R^{-1})_{mm'}|^2=|T_{mm'}|^2, \quad (45)$$

which proves that our result is the same as that of Bloch and Rabi.

[9] E. Wigner, *Gruppentheorie und ihre Anwendung auf die Quantenmechanik der Atmospektren* (Edwards Brothers, Inc., Ann Arbor, 1944), Chap. 15. See especially Eq. (27), p. 180 which corresponds to Eq. (113) here.

3. APPLICATION TO ANOMALOUS ZEEMAN SPLITTING AND TO TRANSITIONS FOR WHICH $(\gamma',j')\neq(\gamma,j)$

In the special case which we considered in Sec. 2, $W_1(\gamma,j,m)$ was given by

$$W_1(\gamma,j,m)=E_0+(g\mu_0H_z/\hbar)m\hbar =E_0+m\hbar a. \quad (46)$$

[10] F. Bloch and I. I. Rabi (reference 3).

Then in the $|(\gamma,j),m\rangle$ representation the diagonal elements of $\mathcal{H}'$ are

$$\langle(\gamma,j),m|\,\mathcal{H}'\,|(\gamma,j),m\rangle = W_1(\gamma,j,m)-m\hbar\omega = E_0(\gamma,j)+m\hbar[a(\gamma,j)-\omega] \tag{47}$$

[$g=g(\gamma,j)$; see reference 8]. At the frequency $\omega=a(\gamma,j)$, all diagonal elements corresponding to (γ,j) will be equal. This frequency is the resonant frequency in the case of a linear Zeeman effect.

We will deal in this section with the case which is, in a sense, the opposite of the above. We will make the assumption that if ω is chosen so that

$$\langle(\gamma,j),m|\,\mathcal{H}'\,|(\gamma,j),m\rangle \sim\langle(\gamma',j'),m'|\,\mathcal{H}'\,|(\gamma',j'),m'\rangle, \tag{48}$$

then at that frequency the difference between these two diagonal elements and any others is large compared to the off-diagonal elements of $\mathcal{H}'$. In other words, we assume that in terms of the diagonal elements of $\mathcal{H}'$ a given level is degenerate with no more than one other level at a given frequency.

A. Simplified Treatment

Before entering into the actual calculations for this case, we will attempt to show by a somewhat simplified discussion of the problem at hand why the particular method of attack used in the following sections was chosen. Certain expressions will be written down here without justification. The justification will appear in the following sections.

Consider the simplest case to which the method can be applied, that in which the interactions of two levels $m=\pm\frac{1}{2}$, are considered. $\mathcal{H}'$ will then take the form

$$\mathcal{H}'=\begin{pmatrix} W_{\frac{1}{2}}-\frac{1}{2}\hbar\omega & b\hbar\alpha \\ b\hbar\bar{\alpha} & W_{-\frac{1}{2}}+\frac{1}{2}\hbar\omega \end{pmatrix}, \tag{49}$$

where b is proportional to the rf amplitude and α, which in this case is most conveniently taken as $\frac{1}{2}$, will in further examples be taken proportional to the matrix element of $(g_1J_{1x}+g_2J_{2x}+\cdots+g_hJ_{hx})$.

Since the secular equation for $\mathcal{H}'$ is quadratic, we may solve it exactly, obtaining the eigenvalues

$$\lambda_\pm=\tfrac{1}{2}[(W_{\frac{1}{2}}-\tfrac{1}{2}\hbar\omega)+(W_{-1}+\tfrac{1}{2}\hbar\omega)] \pm\tfrac{1}{2}[(W_{\frac{1}{2}}-W_{-\frac{1}{2}}-\hbar\omega)^2+4b^2\hbar^2|\alpha|^2]^{\frac{1}{2}} =\tfrac{1}{2}(W_{\frac{1}{2}}+W_{-\frac{1}{2}})\pm\tfrac{1}{2}\hbar[(\omega_0-\omega)^2+4b^2|\alpha|^2]^{\frac{1}{2}}, \tag{50}$$

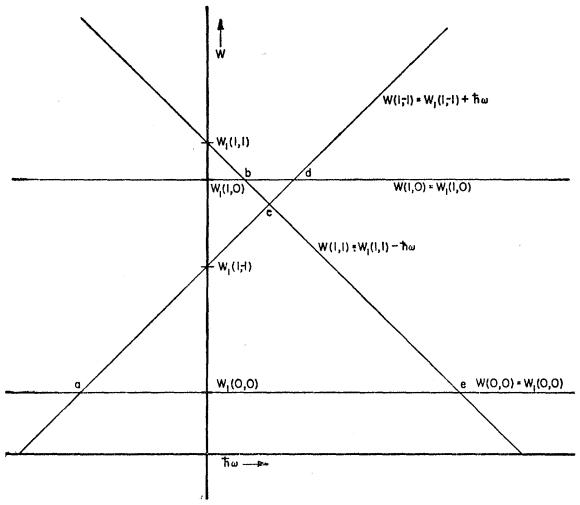

FIG. 2. Diagonal matrix elements as a function of frequency for a typical ordering of the energies $W_1(j,m)$. In this figure $W(j,m)$ is used as an abbreviation for

$$\langle(j),m|\,\mathcal{H}'\,|(j),m\rangle=W_1(j,m)-m\hbar\omega.$$

where the resonant frequency, ω_0, is given by $\hbar\omega_0=W_{\frac{1}{2}}-W_{-\frac{1}{2}}$. The corresponding eigenfunctions will have components

$$\langle\tfrac{1}{2}|\pm\rangle=\frac{b\alpha}{|b\alpha|}\cdot\frac{1}{\sqrt{2}}\left[1\pm\frac{\omega_0-\omega}{[(\omega_0-\omega)^2+4b^2|\alpha|^2]^{\frac{1}{2}}}\right]^{\frac{1}{2}},$$
$$\langle-\tfrac{1}{2}|\pm\rangle=\pm\frac{1}{\sqrt{2}}\left[1\mp\frac{\omega_0-\omega}{[(\omega_0-\omega)^2+4b^2|\alpha|^2]^{\frac{1}{2}}}\right]^{\frac{1}{2}}, \tag{51}$$

so that, according to Eq. (25), the probability of a transition from one of the initial states $|\frac{1}{2}\rangle$ or $|-\frac{1}{2}\rangle$ is

$$P[|\tfrac{1}{2}\rangle\rightarrow]=4|\langle\tfrac{1}{2}|+\rangle|^2|\langle\tfrac{1}{2}|-\rangle|^2\sin^2[(1/2\hbar)(\lambda_+-\lambda_-)\tau] =\frac{4b^2|\alpha|^2}{(\omega_0-\omega)^2+4b^2|\alpha|^2} \times\sin^2\{\tfrac{1}{2}[(\omega_0-\omega)^2+4b^2|\alpha|^2]^{\frac{1}{2}}\tau\} =P[|-\tfrac{1}{2}\rangle\rightarrow]. \tag{52}$$

Now let us look at a more complicated case. If we allow only one value of γ but let j take the two values 1 and 0, $\mathcal{H}'$ will be of the form

$$\mathcal{H}'=\begin{bmatrix} W_1(1,1)-\hbar\omega & b\hbar\alpha(1,1;1,0) & 0 & b\hbar\alpha(1,1;0,0) \\ b\hbar\alpha(1,0;1,1) & W_1(1,0) & b\hbar\alpha(1,0;1,-1) & 0 \\ 0 & b\hbar\alpha(1,-1;1,0) & W_1(1,-1)+\hbar\omega & b\hbar\alpha(1,-1;0,0) \\ b\hbar\alpha(0,0;1,1) & 0 & b\hbar\alpha(0,0;1,-1) & W_1(0,0) \end{bmatrix} \tag{53}$$

in the $|(\gamma,j),m\rangle$ representation. In Fig. 2, we have plotted the diagonal matrix elements of $\mathcal{H}'$ as a function of $\hbar\omega$, for a typical set of values of $W_1(1,1)$, $W_1(1,0)$, $W_1(1,-1)$, and $W_1(0,0)$. It is seen that there are five

crossing points with frequencies given by

$$\begin{aligned}\hbar\omega_a&=W_1(0,0)-W_1(1,-1),\\ \hbar\omega_b&=W_1(1,1)-W_1(1,0),\\ 2\hbar\omega_c&=W_1(1,1)-W_1(1,-1),\\ \hbar\omega_d&=W_1(1,0)-W_1(1,-1),\\ \hbar\omega_e&=W_1(1,1)-W_1(0,0).\end{aligned}\tag{54}$$

It will be noted that the frequencies ω_a, ω_b, ω_d, and ω_e are the frequencies we should expect for the "allowed" transitions of this system. The frequency ω_c corresponds to the "double-jump" transition between the levels (1,1) and (1, −1), so called because the energy $W_1(1,1)-W_1(1,-1)$ is supplied by two equienergetic quanta of energy $\hbar\omega_c$.

Let us now look at what happens in the frequency range $\omega\sim\omega_b$. In this range, the diagonal elements $W_1(0,0)$ and $W_1(1,-1)+\hbar\omega$ are isolated from each other and from the other two diagonal elements, but $W_1(1,1)-\hbar\omega$ is about equal to $W_1(1,0)$. We may therefore expect $\mathcal{H}'$ to have two isolated roots

$$\lambda_{0,0}\sim W_1(0,0),\qquad \lambda_{1,-1}\sim W_1(1,-1)+\hbar\omega,\tag{55}$$

with

$$|\lambda_{0,0}\rangle\sim|(j=0),0\rangle,\qquad |\lambda_{1,-1}\rangle\sim|(j=1),-1\rangle,\tag{56}$$

and two almost degenerate roots

$$\lambda_\pm\sim W_1(1,1)-\hbar\omega\sim W_1(1,0),\tag{57}$$

with

$$|\lambda_\pm\rangle\sim A_\pm|(1),1\rangle+B_\pm|(1),0\rangle.\tag{58}$$

We would then expect to get a good approximation to $\lambda_\pm$ and $|\lambda_\pm\rangle$ by writing a two-by-two matrix in terms of $|(1),1\rangle$ and $|(1),0\rangle$, and including the effects of the other levels only as a perturbation on these. This argument suggests that a good approximation should be given by replacing the Hamiltonian by

$$\mathcal{H}'=\begin{pmatrix}W_1(1,1)+\Delta W(1,1)-\hbar\omega & b\hbar\alpha(1,1;1,0)+\Delta V\\ b\hbar\alpha(1,0;1,1)+\bar{\Delta}\bar{V} & W_1(1,0)+\Delta W(1,0)\end{pmatrix},\tag{59}$$

with

$$\begin{aligned}\Delta W(1,1)&=\frac{|b\hbar\alpha(1,1;0,0)|^2}{[W_1(1,1)-\hbar\omega]-W_1(0,0)}\sim\frac{|b\hbar\alpha(1,1;0,0)|^2}{\hbar(\omega_e-\omega_b)},\\ \Delta W(1,0)&=\frac{|b\hbar\alpha(1,0;1,-1)|^2}{W_1(1,0)-[W_1(1,-1)+\hbar\omega]}\sim\frac{|b\hbar\alpha(1,0;1,-1)|^2}{\hbar(\omega_d-\omega_b)},\\ \Delta V&=\frac{[b\hbar\alpha(1,1;0,0)][b\hbar\alpha(0,0;1,-1)][b\hbar\alpha(1,-1;1,0)]}{\{[W_1(1,1)-\hbar\omega]-W_1(0,0)\}\{[W_1(1,1)-\hbar\omega]-[W_1(1,-1)+\hbar\omega]\}}\\ &\sim\frac{[b\hbar\alpha(1,1;0,0)][b\hbar\alpha(0,0;1,-1)][b\hbar\alpha(1,-1;1,0)]}{\hbar(\omega_e-\omega_b)\cdot 2\hbar(\omega_c-\omega_b)}.\end{aligned}\tag{60}$$

We may carry out the diagonalization of (59) as we did that of (49), obtaining for the dominant terms of $P[|(1),1\rangle\rightarrow]$ and $P[|(1),0\rangle\rightarrow]$

$$\begin{aligned}P[|(1),1\rangle\rightarrow]&\sim P[|(1),0\rangle\rightarrow]\\ &\sim\frac{b^{*2}}{(\omega^*-\omega)^2+b^{*2}}\sin^2\{\tfrac{1}{2}[(\omega^*-\omega)^2+b^{*2}]^{\frac{1}{2}}\tau\},\end{aligned}\tag{61}$$

where

$$\begin{aligned}\hbar\omega^*&=\hbar\omega_b+\Delta W(1,1)-\Delta W(1,0),\\ b^*&=2|b\alpha(1,1;1,0)+(1/\hbar)\Delta V|.\end{aligned}\tag{62}$$

In the case shown in Fig. 2, we would expect this line to be shifted toward lower frequencies since $\Delta W(1,1)$ and $\Delta W(1,0)$ are both positive, with $\Delta W(1,1)$ having a larger energy denominator than $\Delta W(1,0)$.

We may consider the case of $\omega\sim\omega_c$ in the same way. In this case, the diagonal elements $W_1(1,1)-\hbar\omega$ and $W_1(1,-1)+\hbar\omega$ will be almost equal and there will be two eigenstates which are approximated by linear combinations of $|(1),1\rangle$ and $|(1),-1\rangle$. For this case we write, as in Eqs. (59) through (62) above,

$$\mathcal{H}'=\begin{bmatrix}W_1(1,1)\\ \quad+\Delta W(1,1)-\hbar\omega & \Delta V\\ \bar{\Delta}\bar{V} & W_1(1,-1)\\ & \quad+\Delta W(1,-1)+\hbar\omega\end{bmatrix},\tag{63}$$

and

$$\begin{aligned}P[|(1),1\rangle\rightarrow]&\sim P[|(1),-1\rangle\rightarrow]\\ &\sim\frac{b^{*2}}{(\omega^*-\omega)^2+b^{*2}}\sin^2\{(2/2)[(\omega^*-\omega)^2+b^{*2}]^{\frac{1}{2}}\tau]\},\end{aligned}\tag{64}$$

with

$$\Delta W(1,1)=\frac{|b\hbar\alpha(1,1;1,0)|^2}{\hbar(\omega_b-\omega_c)}+\frac{|b\hbar\alpha(1,1;0,0)|^2}{\hbar(\omega_e-\omega_c)},$$

$$\Delta W(1,-1)=\frac{|b\hbar\alpha(1,-1;1,0)|^2}{\hbar(\omega_c-\omega_d)}+\frac{|b\hbar\alpha(1,-1;0,0)|^2}{\hbar(\omega_c-\omega_a)},\tag{65}$$

$$\Delta V=\frac{[b\hbar\alpha(1,1;1,0)][b\hbar\alpha(1,0;1,-1)]}{\hbar(\omega_b-\omega_c)}+\frac{[b\hbar\alpha(1,1;0,0)][b\hbar\alpha(0,0;1,-1)]}{\hbar(\omega_c-\omega_a)},$$

and

$$2\hbar\omega^*=2\hbar\omega_c+\Delta W(1,1)-\Delta W(1,-1),\qquad 2b^*=2|(1/\hbar)\Delta V|.\tag{66}$$

In order to justify the approach taken here, we shall derive in the next section a perturbation method which is based upon an expansion in terms of two nearly degenerate levels.

B. Perturbation Theory[11]

Consider a situation in which two unperturbed levels $|a\rangle$ and $|b\rangle$ are almost degenerate, but their unperturbed energies are quite different from those of any other level. The two perturbed states corresponding to $|a\rangle$ and $|b\rangle$ will then be approximated by a linear combination of $|a\rangle$ and $|b\rangle$. We therefore write

$$|\ \rangle=|a\rangle\langle a|\ \rangle+|b\rangle\langle b|\rangle+\textstyle\sum_n|n\rangle\langle n|\ \rangle,\tag{67}$$

where the index n runs over states other than $|a\rangle$ and $|b\rangle$. (The coefficients $\langle n|\ \rangle$ will then be small.) By substituting (67) into the Schrödinger equation

$$\mathfrak{IC}|\ \rangle=W|\ \rangle,\tag{68}$$

we obtain

$$\langle a|\mathfrak{IC}|a\rangle\langle a|\ \rangle+\langle a|\mathfrak{IC}|b\rangle\langle b|\ \rangle+\textstyle\sum_n\langle a|\mathfrak{IC}|n\rangle\langle n|\ \rangle=W\langle a|\ \rangle,\tag{69a}$$

$$\langle b|\mathfrak{IC}|a\rangle\langle a|\ \rangle+\langle b|\mathfrak{IC}|b\rangle\langle b|\ \rangle+\textstyle\sum_n\langle b|\mathfrak{IC}|n\rangle\langle n|\ \rangle=W\langle b|\ \rangle,\tag{69b}$$

$$\langle n|\mathfrak{IC}|a\rangle\langle a|\ \rangle+\langle n|\mathfrak{IC}|b\rangle\langle b|\ \rangle+\textstyle\sum_{n'}\langle n|\mathfrak{IC}|n'\rangle\langle n'|\ \rangle=W\langle n|\ \rangle.\tag{69c}$$

In principle, Eq. (69c) determines the coefficients $\langle n|\ \rangle$ in terms of $\langle a|\ \rangle$, $\langle b|\ \rangle$, and W, the solution taking the form

$$\langle n|\ \rangle=[W-\langle n|\mathfrak{IC}|n\rangle]^{-1}\times\{A_n(W)\langle a|\ \rangle+B_n(W)\langle b|\ \rangle\}.\tag{70}$$

If we are dealing with a finite number of levels we may solve exactly for the coefficients $A_n(W)$ and $B_n(W)$. If we are dealing with an infinite number of levels, or if the finite number is too large to make an exact solution convenient, we may expand A_n and B_n in terms of the off-diagonal part of $\mathfrak{IC}'$. In any case, we may substitute Eq. (70) into Eq. (69a, b), thereby obtaining

$$\begin{pmatrix}\langle a|\mathfrak{IC}|a\rangle+V_{aa}(W) & V_{ab}(W)\\ V_{ba}(W) & \langle b|\mathfrak{IC}|b\rangle+V_{bb}(W)\end{pmatrix}\begin{pmatrix}\langle a|\ \rangle\\ \langle b|\ \rangle\end{pmatrix}=W\begin{pmatrix}\langle a|\ \rangle\\ \langle b|\ \rangle\end{pmatrix},\tag{71}$$

where

$$V_{aa}(W)=\bar V_{aa}(W)=\sum_n\frac{\langle a|\mathfrak{IC}|n\rangle A_n(W)}{W-\langle n|\mathfrak{IC}|n\rangle},$$

$$V_{ab}(W)=\bar V_{ba}(W)=\langle a|\mathfrak{IC}|b\rangle+\sum_n\frac{\langle a|\mathfrak{IC}|n\rangle B_n(W)}{W-\langle n|\mathfrak{IC}|n\rangle},\tag{72}$$

$$V_{bb}(W)=\bar V_{bb}(W)=\sum_n\frac{\langle b|\mathfrak{IC}|n\rangle B_n(W)}{W-\langle n|\mathfrak{IC}|n\rangle}.$$

Equation (71) provides us with an iterative method for determining the perturbed energies. When we insert an approximate value of W into the left hand side of (71) the problem reduces to that of diagonalizing a two-by-two matrix. This yields us two eigenvalues, $W_\pm$. Now, if we put *one* of these into the left hand side of (71) we obtain *two* roots again, one of which corresponds to the eigenvalue for which we are looking. By selecting the proper root and continuing this process one may obtain as good an approximation as one desires.

The expressions for $W_\pm$, $\langle a|\pm\rangle$, and $\langle b|\pm\rangle$ (with the proper root selected) are

$$W_\pm=\tfrac{1}{2}\{[\langle a|\mathfrak{IC}|a\rangle+\langle b|\mathfrak{IC}|b\rangle]+[V_{aa}(W_\pm)+V_{bb}(W_\pm)]\}\pm\tfrac{1}{2}\{[\delta W(W_\pm)]^2+4|V_{ab}(W_\pm)|^2\}^{\frac{1}{2}},\tag{73}$$

and

$$\langle a|\pm\rangle=\frac{V_{ab}(W_\pm)}{|V_{ab}(W_\pm)|}\cdot\frac{N_\pm}{\sqrt{2}}\times\left[1\pm\frac{\delta W(W_\pm)}{\{[\delta W(W_\pm)]^2+4|V_{ab}(W_\pm)|^2\}^{\frac{1}{2}}}\right]^{\frac{1}{2}},$$

$$\langle b|\pm\rangle=\pm\frac{N_\pm}{\sqrt{2}}\left[1\mp\frac{\delta W(W_\pm)}{\{[\delta W(W_\pm)]^2+4|V_{ab}(W_\pm)|^2\}^{\frac{1}{2}}}\right]^{\frac{1}{2}}\tag{74}$$

[11] For related perturbation methods see J. H. Van Vleck, Phys. Rev. **33**, 467 (1929); O. M. Jordahl, Phys. Rev. **45**, 87 (1934); M. H. L. Pryce, Proc. Phys. Soc. (London) **A63**, 25 (1950); O. M. Jordahl attributes his method to Kramers, Proc. Amsterdam Acad. Sci. **35**, 1272 (1932).

where

$$\delta W(W_\pm)=[\langle a|\,\mathcal{H}|a\rangle+V_{aa}(W_\pm)]]-[\langle b|\,\mathcal{H}|b\rangle+V_{bb}(W_\pm)], \quad (75)$$

and $N_\pm$ is a normalization constant, which is determined by the equation

$$|\langle a|\pm\rangle|^2+|\langle b|\pm\rangle|^2+\sum_n|\langle n|\pm\rangle|^2=1. \quad (76)$$

Once Eq. (73) has been used to obtain an accurate value of $W_\pm$, we may use Eqs. (74) and (70), to get the corresponding eigenstates, $|\pm\rangle$.

C. Transition Probabilities Near a Resonance

The results of the preceding section should enable us, in any particular case, to calculate the transition probability to the desired accuracy. We shall now carry this calculation through to a first approximation for the states $|a\rangle=|(\gamma_a,j_a),m_a\rangle$ and $|b\rangle=|(\gamma_b,j_b),m_b\rangle$.

Put $\mathcal{H}'$ in place of $\mathcal{H}$ in the equations of Sec. 3. B and let W_0 be defined by

$$W_0=\tfrac{1}{2}\{[\langle a|\,\mathcal{H}'|a\rangle+\langle b|\,\mathcal{H}'|b\rangle]+[V_{aa}(W_0)+V_{bb}(W_0)]\}. \quad (77)$$

If we use W_0 as a zeroth approximation to $W_\pm$, Eqs. (73) and (74) now give

$$W_\pm=W_0\pm\tfrac{1}{2}\{[\delta W(W_0)]^2+4|V_{ab}(W_0)|^2\}^{\frac{1}{2}}, \quad (78)$$

and

$$\langle a|\pm\rangle=\frac{V_{ab}(W_0)}{|V_{ab}(W_0)|}\cdot\frac{N_\pm}{\sqrt{2}}\times\left[1\pm\frac{\delta W(W_0)}{\{[\delta W(W_0)]^2+4|V_{ab}(W_0)|^2\}^{\frac{1}{2}}}\right]^{\frac{1}{2}}$$
$$\langle b|\pm\rangle=\pm\frac{N_\pm}{\sqrt{2}}\left[1\mp\frac{\delta W(W_0)}{\{[\delta W(W_0)]^2+4|V_{ab}(W_0)|^2\}^{\frac{1}{2}}}\right]^{\frac{1}{2}}. \quad (79)$$

Then the term in the transition probability which is large near the resonance $a\leftrightarrow b$ is given to this approximation by

$$4|\langle a|+\rangle|^2|\langle a|-\rangle|^2\sin^2(1/2\hbar)(W_+-W_-)\tau \sim|N_+|^2|N_-|^2\frac{4|V_{ab}(W_0)|^2}{[\delta W(W_0)]^2+4|V_{ab}(W_0)|^2}\times\sin^2(1/2\hbar)\{[\delta W(W_0)]^2+4|V_{ab}(W_0)|^2\}^{\frac{1}{2}}\tau. \quad (80)$$

Since

$$\langle a|\,\mathcal{H}'|a\rangle=W_1(\gamma_a,j_a,m_a)-m_a\hbar\omega\equiv W_1(a)-m_a\hbar\omega,$$
$$\langle b|\,\mathcal{H}'|b\rangle=W_1(\gamma_b,j_b,m_b)-m_b\hbar\omega\equiv W_1(b)-m_b\hbar\omega. \quad (81)$$

$\delta W(W_0)$ is given by

$$\delta W(W_0)=(m_a-m_b)\hbar(\omega_{ab}-\omega)+[V_{aa}(\omega_0)-V_{bb}(\omega_0)] \quad (82)$$

where ω_{ab} is defined by

$$(m_a-m_b)\hbar\omega_{ab}=W_1(a)-W_1(b). \quad (83)$$

We must bear in mind that W_0 is a function of ω. Nevertheless, it is clear from the form of Eq. (82) that there is a frequency $\omega^*\sim\omega_{ab}$ such that $\delta W(W_0(\omega^*))=0$. The frequency ω^* is then the corrected resonant frequency and, if we make the approximation that changes in V_{aa}, V_{bb}, and V_{ab} in the vicinity of the resonance may be ignored, the line shape will be given by

$$P[|a\rangle\rightarrow]\sim P[|b\rangle\rightarrow]\sim\frac{|N_+|^2|N_-|^2\cdot4|(1/\hbar)V_{ab}(W_0(\omega^*))|^2}{(m_a-m_b)^2(\omega^*-\omega)^2+4|(1/\hbar)V_{ab}(W_0(\omega^*))|^2}\times\sin^2\tfrac{1}{2}\{(m_a-m_b)^2(\omega^*-\omega)^2+4|(1/\hbar)V_{ab}(W_0(\omega^*))|^2\}^{\frac{1}{2}}\tau. \quad (84)$$

In analogy with Torrey's Eq. (4),[4] we may, omitting the factor $|N_+|^2|N_-|^2\sim1$, put this in the form

$$P[|a\rangle\rightarrow]\sim P[|b\rangle\rightarrow]\sim\frac{b_{ab}^2}{(\nu_{ab}^*-\nu)^2+b_{ab}^2}\times\sin^2\{\pi|m_a-m_b|[(\nu_{ab}^*-\nu)^2+b_{ab}^2]^{\frac{1}{2}}\tau\}, \quad (85)$$

with

$$\nu_{ab}^*=\omega^*/2\pi \quad (86)$$

and

$$b_{ab}=2|V_{ab}|/(|m_a-m_b|\hbar). \quad (87)$$

It may be shown that in the neighborhood of a resonance $|N_+|^2|N_-|^2=1+O(b^2)$ and terms other than (80) in the transition probability are of the order of b^2. It may also be shown that both these corrections are independent of frequency except for fourth-order terms. Thus Eq. (84) should give a good approximation in all cases in which the widths of "allowed" transitions are small compared to their separations.

D. Approximate Values of b_{ab} and ν_{ab}^*

In Sec. 3. B we merely remarked that Eq. (69c) could be solved for the coefficients $\langle n|\ \rangle$ but we did not carry out that solution. In this section we shall obtain the solution to the lowest order in b (the measure of the rf amplitude) and use that solution to get approximate values for the constants b_{ab} and ν_{ab}^* of Eq. (85).

Before carrying this out it is convenient to specify the states $|n\rangle$, $|n'\rangle$, etc., by

$$|n\rangle=|(\gamma,j),m\rangle,\quad|n'\rangle=|(\gamma',j'),m'\rangle,\quad\ldots\quad|n^{(k)}\rangle=|(\gamma^{(k)},j^{(k)}),m^{(k)}\rangle, \quad (88)$$

in analogy with the notation for $|a\rangle$ and $|b\rangle$. We will then define such expressions as ω_{an}, $\omega_{nn'}$, and $W_1(n)$, as we did similar expressions in Eqs. (31) and (33).

Now note that

$$\mathfrak{IC}'=(\mathfrak{IC}_1-\omega J_z)+(\mu_0H_0/\hbar)\times[g_1J_{1x}+g_2J_{2x}+\cdots+g_hJ_{hx}], \quad (13)$$

so that

$$\begin{aligned}&\langle n|\,\mathfrak{IC}'\,|n\rangle=W_1(\gamma,j,m)-m\hbar\omega\equiv W_1(n)-m\hbar\omega,\\&\langle n|\,\mathfrak{IC}'\,|n'\rangle=\langle n|\,(\mu_0H_0/\hbar)\times[g_1J_{1x}+\cdots+g_hJ_{hx}]\,|n'\rangle \quad \text{for } n'\neq n,\end{aligned} \quad (89)$$

where $\langle n|\,(\mu_0H_0/\hbar)[g_1J_{1x}+\cdots+g_hJ_{hx}|n'\rangle$ is zero unless $m=m'\pm1$. Then by suitable definitions of b and $\alpha(n;n')$ we have

$$\begin{aligned}&\langle n|\,\mathfrak{IC}'\,|n\rangle=W_1(n)-m\hbar\omega,\\&\langle n|\,\mathfrak{IC}'\,|n'\rangle=b\hbar\alpha(n;n') \quad \text{for } n'\neq n,\end{aligned} \quad (90)$$

where

$$\alpha(n;n')=0, \quad \text{unless } m=m'\pm1. \quad (91)$$

We shall adopt the convention that b is proportional to the rf amplitude and independent of the states involved, while $\alpha(n;n')$ depends only on the states.

Equation (69c) now becomes

$$[W-\langle n|\,\mathfrak{IC}'\,|n\rangle]\langle n|\ \rangle=b\hbar\alpha(n;a)\langle a|\ \rangle+b\hbar\alpha(n;b)\langle b|\ \rangle+\sum_{m'=m\pm1}b\hbar\alpha(n;n')\langle n'|\ \rangle, \quad (92)$$

where the summation is over states other than $|a\rangle$ and $|b\rangle$. Then, due to the form of Eq. (70), we obtain

$$\begin{aligned}A_n(W)&=b\hbar\alpha(n;a)+\sum_{m'=m\pm1}\frac{b\hbar\alpha(n;n')}{W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle}A_{n'}(W),\\B_n(W)&=b\hbar\alpha(n;b)+\sum_{m'=m\pm1}\frac{b\hbar\alpha(n;n')}{W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle}B_{n'}(W).\end{aligned} \quad (93)$$

We thus note that

$$A_n(W)=\begin{cases}O(b^2) & \text{if } m=m_a\\ O(b^k) & \text{if } |m-m_a|=k\neq0,\end{cases} \quad (94)$$

with similar relations for $B_n(W)$.

We shall approximate $A_n(W)$ and $B_n(W)$ by their lowest order terms:

$$A_n(W)=\begin{cases}b\hbar\alpha(n;a) & \text{for } |m-m_a|=1\\ b\hbar\displaystyle\sum_{|m'-m_a|=1}\frac{[b\hbar\alpha(n;n')][b\hbar\alpha(n';a)]}{W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle} & \text{for } m=m_a\\ b\hbar\displaystyle\sum_{m'=m_a\pm(k-1)}\frac{b\hbar\alpha(n;n')}{W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle}A_{n'}(W) & \text{for } m=m_a\pm k,\ k>1\end{cases} \quad (95a)$$

$$B_n(W)=\begin{cases}b\hbar\alpha(n;b) & \text{for } |m-m_b|=1\\ b\hbar\displaystyle\sum_{|m'-m_b|=1}\frac{[b\hbar\alpha(n;n')][b\hbar\alpha(n';b)]}{W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle} & \text{for } m=m_b\\ b\hbar\displaystyle\sum_{m'=m_b\pm(k-1)}\frac{b\hbar\alpha(n;n')}{W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle}B_{n'}(W). & \text{for } m=m_b\pm k,\quad k>1\end{cases} \quad (95b)$$

The lowest order terms in $V_{aa}(W)$, $V_{bb}(W)$, and $V_{ab}(W)$ are then (see Eq. (72)):

$$V_{aa}(W)=b^2\hbar^2\sum_{|m'-m_a|=1}\frac{|\alpha(n';a)|^2}{W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle}+O(b^4), \quad (96a)$$

$$V_{bb}(W)=b^2\hbar^2\sum_{|m'-m'|=1}\frac{|\alpha(n';b)|^2}{W-\langle n'|\,\mathfrak{IC}\,|n'\rangle}+O(b^4), \quad (96b)$$

$$V_{ab}(W)=\begin{cases}b\hbar\alpha(a;b)+O(b^3) & \text{for } |m_a-m_b|=1\\ b^k\hbar^k\displaystyle\sum_{\substack{m'=m_a\pm1\\ m''=m_a\pm2\\ \cdots\\ m^{(k-1)}=m_a\pm(k-1)}}\frac{\alpha(a;n')\alpha(n';n'')\cdots\alpha(n^{(k-1)};b)}{[W-\langle n'|\,\mathfrak{IC}'\,|n'\rangle][W-\langle n''|\,W'\,|n''\rangle]\cdots[W-\langle n^{(k-1)}|\,\mathfrak{IC}'\,|n^{(k-1)}\rangle]}+O(b^{k+2}) & \text{for } m_b=m_2\pm k,\quad k>1\end{cases} \quad (96c)$$

Then $\delta W(W)$ becomes

$$\begin{aligned}\delta W(W)&=\langle a|\,\mathfrak{IC}'\,|a\rangle-\langle b|\,\mathfrak{IC}'\,|b\rangle+V_{aa}(W)-V_{bb}(W)\\&=[W_1(a)-m_a\hbar\omega]-[W_1(b)-m_b\hbar\omega]+V_{aa}(W)-V_{bb}(W)\\&=(m_a-m_b)\hbar(\omega_{ab}-\omega)+V_{aa}(W)-V_{bb}(W).\end{aligned} \quad (97)$$

Since ω^* is defined as the root of $\delta W(W_0(\omega^*))=0$, we now obtain

$$\begin{aligned}\omega^*&=\omega_{ab}\mp(1/\hbar k)\{V_{aa}(W_0(\omega^*))-V_{bb}(W_0(\omega^*))\}\\&=\omega_{ab}+O(b^2)\\&=\omega_{ab}\mp(1/\hbar k)\{V_{aa}(W_0(\omega_{ab}))-V_{bb}(W_0(\omega_{ab}))\}+O(b^4),\end{aligned} \quad (98)$$

where

$$\pm k=m_b-m_a; \quad k>0. \quad (99)$$

This leads to

$$\begin{aligned}W_0(\omega^*)&=\tfrac{1}{2}[\langle a|\,\mathcal{H}'\,|a\rangle+\langle b|\,\mathcal{H}'\,|b\rangle]\\&\quad+\tfrac{1}{2}[V_{aa}(W_0(\omega^*))+V_{bb}(W_0(\omega^*))]\\&=\tfrac{1}{2}\{[W_1(a)-m_a\hbar\omega^*]\\&\quad+[W_1(b)-m_b\hbar\omega^*]\}+O(b^2)\\&=\tfrac{1}{2}\{[W_1(a)-m_a\hbar\omega_{ab}]\\&\quad+[W_1(b)-m_b\hbar\omega_{ab}]\}+O(b^2)\\&=W_1(a)-m_a\hbar\omega_{ab}+O(b^2)\\&=W_1(b)-m_b\hbar\omega_{ab}+O(b^2).\end{aligned}\tag{100}$$

Then we have

$$\begin{aligned}&W_0(\omega^*)-\langle n'|\,\mathcal{H}'\,|n'\rangle\\&\quad=[W_1(a)-m_a\hbar\omega_{ab}]-[W_1(n')-m'\hbar\omega^*]+O(b^2)\\&\quad=(m_a-m')\hbar(\omega_{an'}-\omega_{ab})+O(b^2)\\&\quad=(m_b-m')\hbar(\omega_{bn'}-\omega_{ab})+O(b^2).\end{aligned}\tag{101}$$

Finally, we obtain

$$\omega^*=\omega_{ab}\mp(b^2/k)\left\{\sum_{m'=m_a-1}\frac{|\alpha(n';a)|^2}{\omega_{an'}-\omega_{ab}}+\sum_{m'=m_a+1}\frac{|\alpha(n';a)|^2}{\omega_{ab}-\omega_{an'}}-\sum_{m'=m_b-1}\frac{|\alpha(n';b)|^2}{\omega_{bn'}-\omega_{ab}}-\sum_{m'=m_b+1}\frac{|\alpha(n';b)|^2}{\omega_{ab}-\omega_{bn'}}\right\}+O(b^4)\tag{102}$$

and

$$V_{ab}(W_0(\omega^*))=\begin{cases}b\hbar\alpha(a;b)+O(b^3) & \text{for } k=1\\ b^k\hbar\displaystyle\sum_{\substack{m'=m_a\pm1\\ m''=m_a\pm2\\ \cdots\\ m^{(k-1)}=m_a\pm(k-1)}}\dfrac{\alpha(a;n')\alpha(n';n'')\cdots\alpha(n^{(k-1)};b)}{(\pm)(\omega_{ab}-\omega_{bn'})(\pm)2(\omega_{ab}-\psi_{bn''})\cdots(\pm)(k-1)(\omega_{ab}-\omega_{bn^{(k-1)}})}+O(b^{k+2}) & \text{for } k>1\end{cases}\tag{103}$$

The above results immediately yield

$$\nu_{ab}{}^*=\nu_{ab}\mp(1/k)(b/2\pi)^2\left\{\sum_{m'=m_a-1}\frac{|\alpha(n';a)|^2}{\nu_{an'}-\nu_{ab}}+\sum_{m'=m_a+1}\frac{|\alpha(n';a)|^2}{\nu_{ab}-\nu_{an'}}-\sum_{m'=m_b-1}\frac{|\alpha(n';b)|^2}{\nu_{bn'}-\nu_{ab}}-\sum_{m'=m_b+1}\frac{|\alpha(n';b)|^2}{\nu_{ab}-\nu_{bn'}}\right\}+O(b^4),\tag{104}$$

and

$$b_{ab}=\begin{cases}2(b/2\pi)\alpha(a;b)+O(b^2) & \text{for } k=1\\ (2/k!)(b/2\pi)^k\left|\displaystyle\sum_{\substack{m'=m_a\pm1\\ m''=m_a\pm2\\ \cdots\\ m^{(k-1)}=m_a\pm(k-1)}}\dfrac{\alpha(a;n')\alpha(n';n'')\cdots\alpha(n^{(k-1)};b)}{(\nu_{ab}-\nu_{bn'})(\nu_{ab}-\nu_{bn''})\cdots(\nu_{ab}-\nu_{bn^{(k-1)}})}\right|+O(b^{k+2}) & \text{for } k>1\end{cases}\tag{105}$$

where

$$\nu_{ab}\equiv(\omega_{ab}/2\pi),\text{ etc.}\tag{106}$$

We have thus obtained values of the constants b_{ab} and $\nu_{ab}{}^*$ which, when substituted into Eq. (84), will give us the approximate line shape. It should be noted that these results are in essential agreement with those of Sec. 3. A.

4. INTEGRATION OVER THE VELOCITY DISTRIBUTION

All the results obtained up to this point refer to an atom or molecule which is in the field for a given length of time, τ. In a molecular beams experiment this time is the transit time of a particle through the rf field and is therefore dependent on the particle velocity.

For a beam of particles which have escaped through a hole in an oven, the velocity distribution is given by

$$dn\propto v^3\exp(-mv^2/2kT)dv.\tag{107}$$

The result of averaging Eq. (25) over the velocity distribution is then

$$\begin{aligned}&\bar{P}[|\,(t=0)\rangle\to]\\&\quad=8(m/2kT)^2\int_0^\infty v^3\exp(-mv^2/2kT)\\&\qquad\times\sum_{\lambda>\lambda'}|\langle(t=0)|\lambda\rangle|^2|\langle(t=0)|\lambda'\rangle|^2\\&\qquad\times\sin^2[(1/2\hbar)(\lambda-\lambda')L/v]dv\\&\quad=8\sum_{\lambda>\lambda'}|\langle(t=0)|\lambda\rangle|^2|\langle(t=0)|\lambda'\rangle|^2\\&\qquad\times\int_0^\infty x^3\exp(-x^2)\sin^2[(1/2\hbar)(\lambda-\lambda')\\&\qquad\times L(m/2kT)^{\frac{1}{2}}/x]dx,\end{aligned}\tag{108}$$

where L is the distance the particle travels in the rf

field. If we define the integral $K(\beta)$ by

$$K(\beta)=\int_0^\infty x^3\exp(-x^2)\sin^2(\beta/2x)dx, \quad (109)$$

we have

$$\bar{P}[|(t=0)\rangle\rightarrow]=8\sum_{\lambda>\lambda'}|\langle(t=0)|\lambda\rangle|^2|\langle(t=0)|\lambda'\rangle|^2 \times K[(1/\hbar)(\lambda-\lambda')(m/2kT)^{\frac{1}{2}}L]. \quad (110)$$

We thus see that whenever our initial assumptions about homogeneity are valid the average over the velocity distribution is given by the function $K(\beta)$. We have used the methods suggested by Torrey[4] to calculate this integral for the full range of β, and the results will be published in a subsequent paper.

5. CONCLUSIONS

We have shown that when both the constant and the oscillating (rotating) magnetic fields are homogeneous, the solution may be reduced to the solution of an eigenvalue problem. This problem has been solved both for the case of a normal Zeeman effect and for the case in which transitions from a given state are isolated from each other.

For a normal Zeeman effect, the result is in agreement with previously known solutions of the problem. Our result [Eq. (41)] has the advantage that it is in a form in which the integral over the velocity distribution may immediately be obtained in terms of a single (soon to be tabulated) function.

In the second case, an approximate solution has been found for the line shape of a resonance due to a multiple-quantum transition. This result, given in Eq. (85), is in the same form as the "Rabi flopping formula" for allowed transition, except for a factor $|m_a-m_b|$ in the argument of the $\sin^2$. Another way of putting this is to say that the transit time, τ, in the Rabi formula is replaced by $|m_a-m_b|\tau$. We must therefore replace the criterion

$$\Delta\nu\tau_0\sim 1 \quad (111)$$

for the natural width of the line, by

$$|m_a-m_b|\Delta\nu\tau_0\sim 1, \quad (112)$$

where τ_0 is the transit time for the most probable velocity in the oven. The consequent narrowing of "double-flop" and "triple-flop" lines has been observed by Kusch.[12]

The detailed comparison of the theory with experiment has been deferred to a future paper.

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APPENDIX. RELATIONSHIP OF THE WIGNER COEFFICIENTS TO THE $T_{mm'}$ OF BLOCH AND RABI

Wigner's formula for the coefficients $\mathfrak{D}^{(j)}(\alpha,\beta,\gamma)_{\mu'\mu}$ is (Eq. 27, reference 9)

$$\mathfrak{D}^{(j)}(\alpha,\beta,\gamma)_{\mu'\mu}=\sum_{\chi=\mathrm{Max}\left\{\begin{smallmatrix}0\\ \mu-\mu'\end{smallmatrix}\right\}}^{\mathrm{Min}\left\{\begin{smallmatrix}j+\mu\\ j-\mu'\end{smallmatrix}\right\}}(-1)^\chi\left\{\frac{[(j+\mu)!(j-\mu)!(j+\mu')!(j-\mu')!]^{\frac{1}{2}}}{(j+\mu-\chi)!(j-\mu'-\chi)!\chi!(\chi+\mu'-\mu)!}\right\} \times\{e^{i\mu'\alpha}\cos^{2j+\mu-\mu'-2\chi}(\beta/2)\sin^{2\chi+\mu'-\mu}(\beta/2)e^{i\mu\gamma}\} \quad (113)$$

so that $\mathfrak{D}^{(\frac{1}{2})}$ is given by

$$\mathfrak{D}^{(\frac{1}{2})}(\alpha,\beta,\gamma)=\begin{pmatrix}e^{i\alpha/2}\cos(\beta/2)e^{i\gamma/2} & e^{i\alpha/2}\sin(\beta/2)e^{-i\gamma/2}\\ -e^{-i\alpha/2}\sin(\beta/2)e^{i\gamma/2} & e^{-i\alpha/2}\cos(\beta/2)e^{-i\gamma/2}\end{pmatrix}. \quad (114)$$

Let us denote the components of this matrix by the letters A, B, C, and D. That is,

$$\mathfrak{D}^{(\frac{1}{2})}(R)=\begin{pmatrix}A & B\\ C & D\end{pmatrix}, \quad (115)$$

where the coefficients must satisfy the conditions

$$C=-B^*, \quad D=A^*, \quad |A|^2+|B|^2=1. \quad (116)$$

We may then see by direct comparison that

$$\mathfrak{D}^{(j)}(R)_{\mu'\mu}=\sum_{\chi=\mathrm{Max}\left\{\begin{smallmatrix}0\\ \mu-\mu'\end{smallmatrix}\right\}}^{\mathrm{Min}\left\{\begin{smallmatrix}j+\mu\\ j-\mu'\end{smallmatrix}\right\}}\left\{\frac{[(j+\mu)!(j-\mu)!(j+\mu')!(j-\mu')!]^{\frac{1}{2}}}{(j+\mu-\chi)!(\chi+\mu'-\mu)!(j-\mu'-\chi)!}\right\}\{A^{j+\mu-\chi}B^{\chi+\mu'-\mu}C^\chi D^{j-\mu'-\chi}\}. \quad (117)$$

[12] P. Kusch, Phys. Rev. **93**, 1022 (1954). Multiple quantum transitions have also been reported in a magnetic resonance experiment by Brossel, Cagnac, and Kastler [J. phys. radium **15**, 6 (1954)] and in electric quadrupole resonance by V. Hughes and L. Grabner [Phys. Rev. **79**, 829 (1950)].

Bloch and Rabi (Eq. 30, reference 3) give

$$T_{mm'}=\sum_{\rho=\mathrm{Max}\left\{\begin{smallmatrix}0\\-(m+m')\end{smallmatrix}\right\}}^{\mathrm{Min}\left\{\begin{smallmatrix}j-m\\j-m'\end{smallmatrix}\right\}}\left\{\frac{[(j+m)!(j-m)!(j+m')!(j-m')!]^{\frac{1}{2}}}{(m+m'+\rho)!(j-m'-\rho)!(j-m-\rho)!\rho!}\right\}\{A^{m+m'+\rho}B^{j-m'-\rho}C^{j-m-\rho}D^{\rho}\},\tag{118}$$

where A, B, C, and D are the corresponding coefficients for $j=\frac{1}{2}$. If we change the summation index to $\chi=j-m-\rho$ we get

$$T_{mm'}=\sum_{\chi=\mathrm{Max}\left\{\begin{smallmatrix}0\\m'-m\end{smallmatrix}\right\}}^{\mathrm{Min}\left\{\begin{smallmatrix}j+m'\\j-m\end{smallmatrix}\right\}}\left\{\frac{[(j+m)!(j-m)!(j+m')!(j-m')!]^{\frac{1}{2}}}{(j+m'-\chi)!(\chi+m-m')!\chi!(j-m-\chi)!}\right\}\{A^{j+m'-\chi}B^{\chi+m-m'}C^{\chi}D^{j-m-\chi}\}$$
$$=D^{(j)}(R)_{mm'},\tag{119}$$

where (Bloch and Rabi, reference 3, page 243)

$$\mathfrak{D}^{(\frac{1}{2})}(R)=\begin{pmatrix}A & B\\ C & D\end{pmatrix}=\begin{bmatrix}\cos\tfrac{1}{2}\lambda t-i\dfrac{\omega-\omega_0}{\lambda}\sin\tfrac{1}{2}\lambda t & i\dfrac{gH}{\lambda}\sin\tfrac{1}{2}\lambda t\\ i\dfrac{gH}{\lambda}\sin\tfrac{1}{2}\lambda t & \cos\tfrac{1}{2}\lambda t+i\dfrac{\omega-\omega_0}{\lambda}\sin\tfrac{1}{2}\lambda t\end{bmatrix}.\tag{120}$$

If we make the substitutions

$$gH=g_J\mu_0H_0/\hbar=b,\quad \omega_0=a,\quad \lambda\equiv[(\omega-\omega_0)^2+(gH)^2]^{\frac{1}{2}}=[(a-\omega)^2+b^2]^{\frac{1}{2}},\tag{121}$$

for the constants in terms of those in Sec. 2, we obtain

$$\mathfrak{D}^{(\frac{1}{2})}(R)=\begin{pmatrix}\cos(\theta/2)+i\cos\varphi\sin(\theta/2) & i\sin\varphi\sin(\theta/2)\\ i\sin\varphi\sin(\theta/2) & \cos(\theta/2)-i\cos\varphi\sin(\theta/2)\end{pmatrix}.\tag{122}$$

Since $B=C$, the $T_{mm'}$ are symmetric and

$$T_{mm'}=T_{m'm}=\mathfrak{D}^{(j)}(R)_{mm'}.\tag{123}$$