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Thomas R. Carver

Bruce Partridge
Haverford College, bpartrid@haverford.edu

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AAPT PHYSICS EDUCATION | AMERICAN JOURNAL of PHYSICS

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Citation: Am. J. Phys. **34**, 339 (1966); doi: 10.1119/1.1972965
View online: http://dx.doi.org/10.1119/1.1972965
View Table of Contents: http://ajp.aapt.org/resource/1/AJPIAS/v34/i4
Published by the American Association of Physics Teachers

Monitoring Operators in Magnetic Resonance and Light Modulation

THOMAS R. CARVER* AND ROBERT B. PARTRIDGE†
The Clarendon Laboratory, Oxford University, Oxford, England
(Received 7 July 1965)

A density matrix description of magnetic resonance is constructed, with detailed application to spin $J=1$ (or greater) particles. Different types of "monitoring" or "measurement" operators employed in atomic beam, induction, and optical experiments are constructed, and used as a compact introductory basis for the description of resonance line shapes and other characteristics of these experiments. Of particular interest are the similarities and unique differences between the optical experiments known by the names "optical pumping," "double resonance," and "light beats" on the one hand, and magnetic induction and atomic beam experiments on the other.

I. INTRODUCTION

THE great experimental activity in the many fields of magnetic resonance and the large amount of original and review literature in these fields extending over a period of several decades would seem to make further general discussion superfluous. However, the more recent optical experiments which have been popularly called "optical pumping," "double resonance," and "light beat" experiments make it particularly interesting to draw some simple distinctions between the different ways in which information about the magnetic dipole resonance of a spin system may be obtained. We are stimulated to make these points because a description of these experiments may be made in a more unified and, in some cases, simpler form than that which appears in the original literature, and also because the introductory and review literature in the field of "double resonance" is still rather limited or not conveniently accessible.[1] It will be seen that optical monitoring has a greater versatility in measuring the detailed development of the state of the system, in principle at least, than does induction resonance.

Our approach is to construct the density matrix $\varrho(t)$, for an ensemble of spin J particles subjected to a constant magnetic field, a steady rotating field, relaxation or damping processes, and regeneration processes which maintain a population difference between the Zeeman levels which are denoted by m. We then construct the monitoring operators $\mathbf{M}$, appropriate to different resonance experiments and discuss the results of these experiments in terms of the expectation value of $\mathbf{M}$, $\langle\mathbf{M}\rangle=\mathrm{trace}(\varrho\mathbf{M})$. We emphasize the specific results for the $J=1$ system because this is the simplest case which shows new effects in addition to those predicted by the classical Bloch resonance model. The new effects arise essentially because there are more than two energy levels. Our principal reason for using the density matrix description is that it forms a compact and complete description of the state of the system from which the reader may draw the desired conclusions by inspection. We are also motivated by a desire to provide a vivid illustration of the use of the density matrix, examples of which are not plentiful in the textbook literature. Most of the conclusions we draw are implicit in the original literature. We do not concern ourselves with the experimental magnitudes or practical advantages of one method compared to another.

II. THE DENSITY MATRIX

We consider a spin J particle with magnetic moment $\gamma\mathbf{J}$ interacting with a constant magnetic field H_0 and a field of magnitude H_1 rotating at a frequency ω in a plane perpendicular to H_0. This H_1 may be one rotating component of an oscillating field. The Hamiltonian is

$$\mathcal{H}=-\gamma\mathbf{J}\cdot\mathbf{H}$$

$$\mathcal{H}=-\gamma\mathbf{J}\cdot(H_0\hat{k}+H_1\cos\omega t\hat{\imath}+H_1\sin\omega t\hat{\jmath}). \quad (1)$$

* On a John Simon Guggenheim Foundation Fellowship and sabbatical leave from Princeton University, 1964–65.
† Present address: Palmer Physical Laboratory, Princeton University, Princeton, New Jersey.
[1] C. Cohen-Tannoudji, thesis, University of Paris, 1962 [published in Ann. Phys. (Paris) **7**, 423 (1962)]. This remains the definitive study of the optical pumping cycle and the theory of double-resonance phenomena.

We separate the Hamiltonian into time independent and dependent parts,

$$\mathcal{H}_0=\omega_0 J_z \quad \text{and} \quad \mathcal{H}_1=b(J_x\cos\omega t+J_y\sin\omega t), \tag{2}$$

where we define $b=-\gamma H_1$ and $\omega_0=-\gamma H_0$.

We wish to solve the equation of motion of the density matrix[11]

$$\dot{\varrho}=\frac{i}{\hbar}[\varrho,\ \mathcal{H}_0+\mathcal{H}_1], \tag{3}$$

where the square brackets indicate the quantum mechanical commutation operation. Noting that the time-independent Hamiltonian $\mathcal{H}_1'$ may be formed by the transformation

$$\mathcal{H}_1'=e^{(i/\hbar)J_z\omega t}\mathcal{H}_1(t)e^{-(i/\hbar)J_z\omega t}=bJ_x, \tag{4}$$

we attempt a solution in the rotating frame[2] of Fig. 1,

$$\varrho(t)=e^{-(i/\hbar)J_z\omega t}\varrho'(t)e^{(i/\hbar)J_z\omega t}. \tag{5}$$

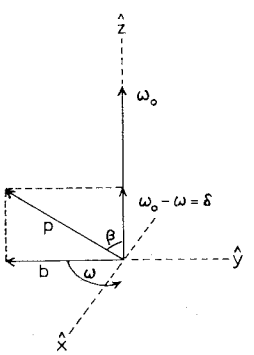

FIG. 1.

Equation (4) may be obvious to those who see that the exponential operator is an operator of rotation around the z axis at frequency ω. It may be shown in detail by considering a particular value of J, or it may be proved (see Ref. 6, p. 25) by defining the inverse transformation to (4),

$$e^{-(i/\hbar)J_z\omega t}J_x e^{(i/\hbar)J_z\omega t}=f(t), \tag{4'}$$

and showing that $f(t)$ obeys the familiar equation

$$d^2f/dt^2+\omega^2 f=0, \tag{4''}$$

whose solution, of course, is $f(t)=J_x\cos\omega t+J_y\sin\omega t$. When (5) is substituted in (3) it gives

$$\dot{\varrho}'(t)=\frac{i}{\hbar}[\varrho'(t),\ J_z(\omega_0-\omega)+bJ_x], \tag{6}$$

where we must make use of the fact that J_z and $\exp(J_z)$ commute. Since the Hamiltonian which we define as

$$\mathcal{H}'=J_z(\omega_0-\omega)+bJ_x=J_z\delta+J_x b \tag{7}$$

is time independent, the formal solution of (6) is

$$\varrho'(t)=e^{-(i/\hbar)\mathcal{H}'(t-t_0)}\varrho_0'(t_0)e^{(i/\hbar)\mathcal{H}'(t-t_0)} \tag{8}$$

and the formal solution in the nonrotating system is

$$\varrho(t)=e^{-(i/\hbar)J_z\omega t}e^{-(i/\hbar)\mathcal{H}'(t-t_0)}\varrho_0'(t_0)\times e^{(i/\hbar)\mathcal{H}'(t-t_0)}e^{(i/\hbar)J_z\omega t}. \tag{9}$$

We must also make the transformation of $\varrho_0'(t_0)$ in (9) to the nonrotating frame,

$$\varrho_0'(t_0)=e^{(i/\hbar)J_z\omega t_0}\varrho_0(t_0)e^{-(i/\hbar)J_z\omega t_0} \tag{10}$$

although this is not necessary if the initial density matrix in the nonrotating frame $\varrho_0(t_0)$ does not have off diagonal elements. Equation (9) is not a convenient form because $\mathcal{H}'$ is not diagonal even though it is time independent. If a time independent unitary transformation can be found to make $\mathcal{H}'$ diagonal in the usual manner such that

$$R^{-1}\mathcal{H}'R=\mathcal{H}_d', \tag{11}$$

it is quickly shown by series expansion[3] of the exponential containing $\mathcal{H}'$ that the identity

$$e^{\pm(i/\hbar)\mathcal{H}'t}\equiv Re^{\pm(i/\hbar)R^{-1}\mathcal{H}'Rt}R^{-1} \tag{11'}$$

permits (9) and (10) to be written as

$$\varrho(t)=e^{-(i/\hbar)J_z\omega t}Re^{-(i/\hbar)\mathcal{H}_d'(t-t_0)}R^{-1}e^{(i/\hbar)J_z\omega t_0}\times\varrho_0(t_0)e^{-(i/\hbar)J_z\omega t_0}Re^{(i/\hbar)\mathcal{H}_d'(t-t_0)}R^{-1}e^{(i/\hbar)J_z\omega t}. \tag{12}$$

By defining $\cos\beta=\delta/p$ and $\sin\beta=b/p$, where

$$p^2=\delta^2+b^2 \quad \text{and} \quad \delta=\omega_0-\omega, \tag{13}$$

so that we may write

$$\mathcal{H}'=J_z\delta+J_x b=(J_z\cos\beta+J_x\sin\beta)p, \tag{14}$$

it can be seen that the necessary transformation to diagonalize $\mathcal{H}'$ in this particular case is the rotation around the y axis through the angle β, which restores the system to the z axis. The R is the rotation operator[4] $D^{(J)}(0,-\beta,0)$ which gives

$$R^{-1}(J_z\cos\beta+J_x\sin\beta)pR=J_z p=\mathcal{H}_d'. \tag{15}$$

At this point it may be helpful to note that, working outward from the initial density matrix $\varrho_0(t_0)$ in Eq. (12), there are a series of unitary

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

[3] W. Franzen and M. Alam, Phys. Rev. **133**, A460 (1964).

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

rotations: to a rotating frame in which the Hamiltonian is made time independent, to a constant tilted frame in which the Hamiltonian is diagonal and may be expressed in terms of simple eigenvalues of $\mathcal{H}_d'$, and back again to the original frame.

By writing the eigenvalues of $J_z\omega$ as $(m, m', m_0,$ or $m_0')\hbar\omega$ and the eigenvalues of $J_z p$ as $(n$ or $n')\hbar p$, the individual elements of the density matrix (12) may be written

$$\rho_{m,m'}(t) = \sum_{\substack{n,n' \\ m_0,m_0'}} e^{-i(m-m'-m_0+m_0')\omega t} R_{m,n} R_{n,m_0} R_{m',n'} R_{n',m_0'} \times e^{-i[(m_0-m_0')\omega+(n-n')p](t-t_0)} \rho_{m_0,m_0'}(t_0). \quad (16)$$

The R_{mn} are the individual matrix elements of the rotation operator and are the same as the $\langle m|\mu\rangle$ of Dodd and Series. We have attempted to follow the notation of Dodd and Series,[5] but we have defined ω_0 as the Larmor frequency and ω as the radio frequency as in slightly more conventional usage.[6] The result (16) is the density matrix equivalent of well-known wavefunction solutions.[2,3,5,7]

The expression (16) is directly useful in atomic beam resonance because there is usually no damping and the time $t-t_0$ represents the transit time of the particle through the region in which the rotating or oscillating field acts on the particle. However, to obtain a practical experimental line shape it is necessary to average the expression over various transit times determined by the velocity distribution in the beam, and perhaps over variations of field strength as well.

In the steady-state resonance of the usual "bulk" sample we must include the effects of relaxation or damping, and of population regeneration. It is beyond the scope and the necessity of this discussion to describe the relaxation as anything other than a uniform decay of all the elements of the density matrix to a uniform distribution, the equation for which is

$$\dot{\varrho}(t) = -\Gamma[\varrho(t) - \mathbf{I}], \quad (17)$$

where $\mathbf{I}$ is the unit or identity matrix. The solution of (17) is

$$\varrho(t) = e^{-\Gamma(t-t_0)}\varrho_0(t_0) + \mathbf{I}. \quad (18)$$

This damping may be caused by radiation which is absorbed or emitted, or by collisions, and we do not wish to distinguish between the damping of diagonal elements (T_1) and the damping of off diagonal elements (T_2) although it may be done.[6] In resonance jargon, $T_1 = T_2$.

We also wish to account for processes of population regeneration in the form

$$\dot{\varrho}(t) = W\varrho^0, \quad (19)$$

where W represents the rate at which particles are steadily supplied to the states of an ensemble in a constant configuration ϱ^0 by means of optical pumping processes, state selectors or thermal processes. Examples are discussed later, but the process of supplying a $J=1$ system with particles in the $m_J=1$ state (polarization), for example, is represented by

$$\varrho_0(t_0) = \varrho^0 = \begin{vmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{vmatrix} \text{ for all } t_0. \quad (20)$$

These steady-state processes may be included by the integral superposition[1,8] of solutions (16) for all t_0 such that $0 < t-t_0 < \infty$, with the damping factor of (18) included, and with the constant ϱ^0 substituted for $\varrho_0(t_0)$. From (16) we can see that the relevant integral is

$$W\varrho^0 \int_0^\infty e^{-i(\Omega-i\Gamma)(t-t_0)} d(t-t_0) = \frac{W\varrho^0}{\Gamma+i\Omega}, \quad (21)$$

where $\Omega = (m_0-m_0')\omega + (n-n')p$. The steady-state system is then described by

$$\rho_{m,m'}(t) = \sum_{\substack{n,n' \\ m_0,m_0'}} e^{-i(m-m'-m_0+m_0')\omega t} W\rho^0{}_{m_0,m_0'} \times \frac{R_{m,n}R_{n,m_0}R_{m',n'}R_{n',m_0'}}{\Gamma+i[(m_0-m_0')\omega+(n-n')p]}. \quad (22)$$

It can be seen that considerable simplification results through the omission of the $m_0-m'_0$ terms in the usual case when ϱ^0 does not contain off diagonal elements.

[5] J. N. Dodd and G. W. Series, Proc. Roy. Soc. (London) **A263**, 353 (1961).

[6] C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963).

[7] H. Salwen, Phys. Rev. **99**, 1274 (1955).

[8] J. P. Barrat, Proc. Roy. Soc. (London) **A263**, 371 (1961).

III. THE DENSITY MATRIX FOR $J=\frac{1}{2}$ AND 1

The evaluation of (22) is somewhat tedious for spins greater than $\frac{1}{2}$ because of the presence of the denominator which is different for each element. It is convenient to make a table of the products of the rotation elements that have a common n, that is, $R_{m,n}R_{n,m_0}$. The R matrix for spin $\frac{1}{2}$ is[4]

$$R^{(\frac{1}{2})}=\begin{vmatrix}\cos\frac{1}{2}\beta & -\sin\frac{1}{2}\beta\\ \sin\frac{1}{2}\beta & \cos\frac{1}{2}\beta\end{vmatrix}, \tag{23}$$

and for spin 1 is

$$R^{(1)}=\begin{vmatrix}\cos^2\frac{1}{2}\beta & -\sqrt{2}\sin\frac{1}{2}\beta\cos\frac{1}{2}\beta & \sin^2\frac{1}{2}\beta\\ \sqrt{2}\sin\frac{1}{2}\beta\cos\frac{1}{2}\beta & \cos^2\frac{1}{2}\beta-\sin^2\frac{1}{2}\beta & -\sqrt{2}\sin\frac{1}{2}\beta\cos\frac{1}{2}\beta\\ \sin^2\frac{1}{2}\beta & \sqrt{2}\sin\frac{1}{2}\beta\cos\frac{1}{2}\beta & \cos^2\frac{1}{2}\beta\end{vmatrix}. \tag{24}$$

It is useful to remind the reader that $R^{(1)}$ may be found from $R^{(\frac{1}{2})}$ by the operation

$$R^{(j+j')+(j-j')}=U^{-1}R^{(j)}\otimes R^{(j')}U$$

and the extraction of the appropriate submatrix for $J=j+j'$ or $J=j-j'$. In this case $J=1$ and $j=j'=\frac{1}{2}$. $\otimes$ represents an outer product, and the U's are coupling matrices whose elements are the Wigner vector coupling coefficients.[9] In this case

$$U=\begin{array}{c|cccc} & 1,1 & 1,0 & 0,0 & 1,-1\\ \hline \frac{1}{2},\frac{1}{2} & 1 & 0 & 0 & 0\\ \frac{1}{2},-\frac{1}{2} & 0 & 2^{-\frac{1}{2}} & 2^{-\frac{1}{2}} & 0\\ -\frac{1}{2},\frac{1}{2} & 0 & 2^{-\frac{1}{2}} & -2^{-\frac{1}{2}} & 0\\ -\frac{1}{2},-\frac{1}{2} & 0 & 0 & 0 & 1\end{array}. \tag{26}$$

The terms of the density matrix, (16) or (22), found element by element, may then be simplified by frequent use of the identities

$$\cos^2\tfrac{1}{2}\beta+\sin^2\tfrac{1}{2}\beta=1;\quad \cos^2\tfrac{1}{2}\beta-\sin^2\tfrac{1}{2}\beta=\delta/p$$
$$2\cos\tfrac{1}{2}\beta\sin\tfrac{1}{2}\beta=b/p;\quad \cos^4\tfrac{1}{2}\beta-\sin^4\tfrac{1}{2}\beta=\delta/p. \tag{27}$$

For simplicity and versatility we give the results for the particular case $\varrho_+(t)$ in which we have supplied polarization to the ensemble in the form given by (20) or its spin $\frac{1}{2}$ equivalent. Other cases may be found by superposition.

The result for $J=\frac{1}{2}$ is

$$\varrho_+^{\frac{1}{2}}(t)=\frac{W}{\Gamma}\begin{vmatrix}1-\frac{1}{2}P & \frac{1}{2}(\chi'+i\chi'')e^{-i\omega t}\\ \frac{1}{2}(\chi'-i\chi'')e^{i\omega t} & \frac{1}{2}P\end{vmatrix}, \tag{28}$$

where

$$P=b^2/\Delta_1;\quad \Delta_1=(\Gamma^2+\delta^2+b^2);$$
$$\chi'=b\delta/\Delta_1;\quad \chi''=b\Gamma/\Delta_1. \tag{28'}$$

Except for a constant, χ' and χ'' are the same as the Bloch functions[10]; hence our notation. The result for $J=1$ is $\varrho_+^1(t)=$

$$\frac{W}{\Gamma}\begin{vmatrix}1-A-F/2 & 2^{-\frac{1}{2}}(X+iY)e^{-i\omega t} & 2^{-1}(D+iE)e^{-i2\omega t}\\ 2^{-\frac{1}{2}}(X-iY)e^{i\omega t} & A & 2^{-\frac{1}{2}}(X'+iY')e^{-i\omega t}\\ 2^{-1}(D-iE)e^{i2\omega t} & 2^{-\frac{1}{2}}(X'-iY')e^{i\omega t} & F/2\end{vmatrix}, \tag{29}$$

where

$$X=b\delta(4\delta^2+b^2+\Gamma^2)/\Delta_2$$
$$Y=b\Gamma(\Gamma^2+4\delta^2+5b^2/2)/\Delta_2$$
$$X'=3\delta b^3/\Delta_2$$
$$Y'=3\Gamma b^3/2\Delta_2$$
$$X-X'=B/2;\quad X+X'=\chi'$$
$$Y-Y'=C/2;\quad Y+Y'=\chi'',$$

and where the resonance functions, A through F, of $\omega_0-\omega$ are defined to be the same as those of Dodd and Series.[5] We define them below for convenience, but the reader is directed to Refs. 22 and 26 for plots of these functions

$$A=b^2(4\delta^2+b^2+\Gamma^2)/\Delta_2,$$
$$B=2b\delta(4\delta^2-2b^2+\Gamma^2)/\Delta_2,$$
$$C=2b\Gamma(4\delta^2+b^2+\Gamma^2)/\Delta_2,$$
$$D=b^2(2\delta^2-b^2-\Gamma^2)/\Delta_2,$$
$$E=3b^2\Gamma\delta/\Delta_2,$$
$$F=3b^4/\Delta_2, \tag{29'}$$

[9] V. Heine, *Group Theory in Quantum Mechanics* (Pergamon Press, Inc., New York, 1960).

[10] See Ref. 6; also, G. E. Pake, Am. J. Phys. **18**, 438, 473 (1950), or A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon Press, Oxford, 1961).

and the denominator

$$\Delta_2=(\Gamma^2+b^2+\delta^2)(\Gamma^2+4b^2+4\delta^2).$$

The factor W/Γ appearing as a multiplier of these density matrices is a ratio of population regeneration maintained in the sample to relaxation, and is a measure of the actual excess population difference from the unpolarized sample represented by **I**.

We also write below the density matrix (16) for spin 1 for a fixed $t-t_0=\tau$ which represents the action of a radio-frequency pulse of duration τ or the passage of a mono-energetic particle through an atomic beam machine, where there is no damping and where $m_J=1$ initial state selection (20) has been made.

$$\rho_{1,1}=1-\left[\frac{b^2\delta^2}{p^4}+\frac{b^4}{2p^4}\right](1-\cos p\tau)$$
$$-\frac{b^4}{8p^4}(1-\cos 2p\tau),$$

$$\rho_{0,0}=\frac{b^2\delta^2}{p^4}(1-\cos p\tau)+\frac{b^4}{4p^4}(1-\cos 2p\tau),$$

$$\rho_{-1,-1}=\frac{b^4}{2p^4}(1-\cos p\tau)-\frac{b^4}{8p^4}(1-\cos 2p\tau),$$

$$\rho_{1,0}=\rho_{0,1}{}^*=\sqrt{2}e^{-i\omega t}\left\{\frac{1}{2}\left[\frac{b\delta^3}{p^4}(1-\cos p\tau)\right.\right.$$
$$\left.+i\frac{b\delta^2}{p^3}\sin p\tau\right]+\frac{1}{4}\left[i\frac{b^3}{p^3}\sin p\tau\right]$$
$$\left.+\frac{1}{8}\left[\frac{b^3\delta}{p^4}(1-\cos 2p\tau)+i\frac{b^3}{p^3}\sin 2p\tau\right]\right\},$$

$$\rho_{0,1}=\rho_{1,0}{}^*=\sqrt{2}e^{-i\omega t}\left\{\frac{1}{4}\left[\frac{b^3\delta}{p^4}(1-\cos p\tau)\right]\right.$$
$$+\frac{1}{4}\left[\frac{b^3\delta}{p^4}(1-\cos p\tau)+i\frac{b^3}{p^3}\sin p\tau\right]$$
$$\left.-\frac{1}{8}\left[\frac{b^3\delta}{p^4}(1-\cos 2p\tau)+i\frac{b^3}{p^3}\sin 2p\tau\right]\right\},$$

$$\rho_{-1,1}=\rho_{1,-1}{}^*=e^{-2i\omega t}$$
$$\times\left\{\frac{1}{2}\left[\frac{b^2\delta^2}{p^4}(1-\cos p\tau)+i\frac{b^2\delta}{p^3}\sin p\tau\right]\right.$$
$$-\frac{1}{4}\left[\frac{b^2\delta^2}{p^4}(1-\cos 2p\tau)+i\frac{b^2\delta}{p^3}\sin 2p\tau\right]$$
$$\left.-\frac{1}{8}\left[\frac{b^4}{p^4}(1-\cos 2p\tau)\right]\right\}. \quad (30)$$

Especially for the student of magnetic resonance who has familiarized himself only with the classical magnetic resonance solutions, a close study of (30) reveals a number of interesting features about the quantum mechanical evolution of the system: in particular, the 2ω and $2p$ components of frequency in the motion[3] should be noted.

IV. MONITORING OPERATORS AND CORRESPONDING EXPERIMENTS

By examining these density matrices, the characteristics and line shapes of several different magnetic resonance experiments may be seen. In each of these experiments a measurement is made which we may represent by some physical monitoring operator **M**, and the result is the expectation value of **M** which is determined from the well-known relation[11]

$$\langle\mathbf{M}\rangle=\mathrm{Trace}(\varrho\mathbf{M})=\sum_{m,m'}\rho_{m,m'}(t)\mathrm{M}_{m',m}. \quad (31)$$

An advantage of this point of view is that the results may be seen by inspection of (29) or (30) when the rather simple form of **M** is known.[12]

A. Atomic Beams

The state selector in the atomic beam experiment[13] is represented by the simplest monitoring

[11] See, for example, Ref. 6 or R. H. Dicke and J. P. Wittke, *Introduction to Quantum Mechanics* (Addison-Wesley, Reading, Mass., 1960).

[12] Since (31) may also be written as

$$\sum_{m,m'}\rho_{m,m'}\mathrm{M}_{m,m'},$$

the process of taking the trace by inspection is equivalent to mentally superposing **M** on ϱ, multiplying the superposed elements and then adding the diagonal, nearest off-diagonal and farthest off-diagonal element products. In this application, these separate sums have different time dependence, 0ω, 1ω, and 2ω, respectively.

[13] We discuss this type of experiment in a limited way only for purposes of comparison. See N. F. Ramsey, *Molecular Beams* (Clarendon Press, Oxford, 1956).

operator. In such experiments a beam of particles is state selected by an inhomogeneous polarizing magnet whose effect is represented by ϱ^0. The beam passes through the static and rotating field region and develops in a manner described by $\varrho(t)$, and finally passes through an analyzing magnet similar to the polarizer whose effect is represented by **M**. Since the force on the particle in the inhomogeneous magnet is

$$F_z=\gamma(dH_z/dz)J_z, \tag{42}$$

and since a slit allows a particular deflection path of the particle, depending on the eigenvalue of J_z, to reach the detector, it is obvious that the **M** operator is represented by elements lying along the m_J diagonal positions of a matrix. A slit so placed as to allow the particle to reach the detector after being "flipped in" to the 0 state from the $m_J=1$ state, for example, is represented by

$$\mathbf{M}_-=K\begin{vmatrix}0&0&0\\0&1&0\\0&0&0\end{vmatrix}. \tag{33}$$

The line-shape function which would be observed would be given by the $\rho_{0,0}$ element of (30), after suitable averaging over an experimental velocity distribution of the particles. Similarly a complete spin flip would be represented by

$$\mathbf{M}_{--}=K\begin{vmatrix}0&0&0\\0&0&0\\0&0&1\end{vmatrix}. \tag{34}$$

We do not consider a constant multiplier of **M** which determines the absolute amplitude of the signal, and which depends on such experimental parameters as the beam intensity, slit widths or detector sensitivity; it is written as K, and does not affect the form of **M** either here or in the forthcoming examples.

B. Magnetic Induction

In this well-known type of magnetic resonance experiment,[10] the sample is placed within a coil or cavity tuned to the frequency ω; the same coil or cavity may be used to supply the H_1 field. The precessing magnetic moment of the sample induces a signal voltage in the coil which, from Faraday's law, is proportional to the rate of change of magnetization. This in turn is proportional to the operator J. Since J_z is diagonal and cannot "pick out" the time varying off-diagonal elements of $\varrho(t)$, it is well known that the coil must be oriented perpendicular to the H_0 or z axis, and the appropriate monitoring operator has the form of J_x or J_y. In the case of $J=1$

$$J_x=\mathbf{M}=K\begin{vmatrix}0&1&0\\1&0&1\\0&1&0\end{vmatrix}. \tag{35}$$

Performing the trace operation (31) using (35) and (29) the signal from the induction experiment is

$$S=K[(X+X')\cos\omega t+(Y+Y')\sin\omega t], \tag{36}$$

or from the properties of the functions defined in (29)

$$S=K(\chi'\cos\omega t+\chi''\sin\omega t). \tag{37}$$

The neglected factor K in this expression contains the Q of the cavity, the γ as defined in (2), and the density, susceptibility, and damping of the sample. An identical result holds for $J=\frac{1}{2}$. Carried out in this way the result (37) might be said to be a "Majorana" derivation rather than a "classical" derivation of the Bloch[10] resonance signal.

That the Bloch functions describe the resonance no matter what the value of J is well known through the fact that they are solutions of the precession equations

$$\dot{\mathbf{J}}=-\gamma(\mathbf{J}\times\mathbf{H})+\text{damping terms}, \tag{38}$$

which are identical to the classical equations and are independent of J. We emphasize the point, for comparison with other monitoring operators, that it is the special form of J_x that produces this result. This shows that such an operator has limitations in its ability to "pick out" the elements of the density matrix.

To avoid confusion it should be pointed out that the sample polarization in a nuclear resonance experiment is usually brought about by thermal processes represented in the high-temperature case by an initial density matrix

$$\varrho^0{}_{\text{thermal}}=\begin{vmatrix}1&0&0\\0&0&0\\0&0&-1\end{vmatrix} \tag{39}$$

instead of $\varrho_+{}^0$. However, the reader may check at once by the obvious superposition of (29) implied by (39) that this does not change the conclusion

(37). In thermal processes the factors W and Γ are equal, and the actual excess population difference is determined by the Boltzmann factor. It may be of some surprise, however, that in the "saturation" limit of high radio-frequency fields ($b \rightarrow \infty$) the populations of the $m=1, 0, -1$ states are in the ratio of 3/8, 1/4, 3/8 in the case where $\varrho_+{}^0$ represents the population regeneration, even though they are equalized by "saturation" when thermal regeneration (39) is present. Briefly, a spin system populated in a nonthermal manner is not saturated to equal random populations, represented by the unit density matrix **I**, even when transitions are allowed between all the states.

C. Optical Monitoring Operators

In optical double-resonance experiments a bulk sample of particles, usually a vapor with suitable spectroscopic properties, is polarized by an optical pumping cycle (to be explained by example later) represented by a particular ϱ^0. It is then magnetically driven by the field H_1. The monitoring process consists of observing the intensity of absorbed or emitted light which connects the group of m levels we have described with another spectroscopically accessible level having m_J states labeled by μ. The light itself has a polarization state represented by the vector **e**, which describes the direction and relative phase of the electric field component of the light. The monitoring beam for absorption experiments is usually derived from a discharge lamp, containing the same element as the sample, which emits an adequately strong line in electric dipole transition. After passing through the sample the partially scattered or absorbed beam is detected by a photodetector, which may either be sensitive only to quasistatic variations as the static field H_0 is varied through the resonance value $\delta=0$, or may be tuned to the fundamental or harmonics of ω, and followed by a phase sensitive amplifier. In the latter case the experiment is sensitive to "light-beat" effects.

It is easy to find the form of the optical monitoring operator for experiments of this type from considerations of the semiclassical correspondence and by the application of the Wigner–Eckart theorem. The reader should be cautioned that there are a number of subtle points concerning the lifetime of the excited states and the spectral distribution of the light which are discussed in the original literature.[5,1,8] These effects happily do not concern us in these examples if we consider that the spectral distribution of the light beam is sufficiently broad to "cover" or overlap the group of states labeled by m or μ, and this is the usual case because of the fact that the spectral distribution from the lamp is always as broad as, and usually broader than, that of the absorbing sample.

In the semiclassical model of absorption, the electric field of the incident light induces an electric dipole moment of the atom which is proportional to the field and is represented by a dipole matrix element. The absorption on which our monitoring operator depends is proportional to the quantum mechanical transition rate, which is well known to be dependent on the square of the field or the square of the dipole matrix element. The form of the monitoring operator is

$$\mathbf{M}=K\sum_{\mu}\langle m|\mathbf{e}\cdot\mathbf{P}|\mu\rangle\langle\mu|\mathbf{e}^*\cdot\mathbf{P}|m'\rangle, \qquad (40)$$

where the individual brackets are the matrix elements of the electric dipole operator **P** or $e\mathbf{r}$ appropriate to the polarization **e** of the incident light, taken between the states m which are monitored, and a final state or states μ. This form for **M** is applicable not only to absorption but also to emission because of the fact that both processes are proportional to the square of the electric dipole matrix element even though they differ in magnitude by the ratio of the Einstein B and A coefficients.

To evaluate **M** we make use of the matrix elements[14] written below for reference and illustration.

$$\begin{aligned}
&\langle n,j,m|P|n',\ j+1,\ m\pm1\rangle=\mp P_1(n,n',j,j')\\
&\qquad\cdot\tfrac{1}{2}[(j\pm m+1)(j\pm m+2)]^{\frac{1}{2}}\cdot(\hat{x}\pm i\hat{y})\\
&\langle j,m|P|j+1,\ m\rangle=P_1\cdot[(j+1)^2-m^2]^{\frac{1}{2}}\cdot\hat{z}\\
&\langle j,m|P|j,\ m\pm1\rangle=P_0\cdot\tfrac{1}{2}[(j\mp m)(j\pm m+1)]^{\frac{1}{2}}\\
&\qquad\cdot(\hat{x}\pm\hat{y}) \qquad (41)\\
&\langle j,m|P|j,m\rangle=P_0\cdot m\cdot\hat{z}\\
&\langle j,m|P|j-1,\ m\pm1\rangle=\pm P_{-1}\\
&\qquad\cdot\tfrac{1}{2}[(j\mp m)(j\mp m-1)]^{\frac{1}{2}}\cdot(\hat{x}\pm i\hat{y})\\
&\langle j,m|P|j-1,\ m\rangle=P_{-1}[j^2-m^2]^{\frac{1}{2}}\cdot\hat{z}.
\end{aligned}$$

[14] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, London, 1951).

These elements consist of a reduced matrix element depending on specific spectroscopic information involving the principal quantum number (shown in the first example and omitted from the notation in the remainder) and the spectroscopic configuration. This reduced matrix element is multiplied by a Clebsch–Gordon coefficient depending only on the angular momentum. This separation is an example of the Wigner–Eckart theorem.[15] It is convenient because we are concerned with transitions between a group of m levels and another group of μ levels, and can therefore absorb the reduced matrix element in the K constant and *find the form of* $\mathbf{M}$ *from the angular momentum coefficients alone.*[16]

To see the physical significance of $\mathbf{M}$, we note that the diagonal elements ($m=m'$) represent the sum of the squares of the dipole matrix elements over all final states μ, and physically represent the sum of all the transitions from each state labeled by m. The off-diagonal elements represent interference products of the components of optical radiation arising from a superposition of m and m' when these states are coupled by magnetic resonance. As illustrated in Fig. 2, these off diagonal elements represent the two interfering paths (a) and (b) which are taken by the system as it passes from an initial state m_0 to μ through the intermediate states m and m'. The $\mathbf{M}$ operator we have written is identical to the G*·G of Dodd and Series.

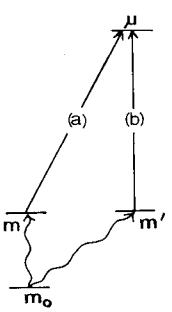

FIG. 2.

D. The Dehmelt Experiment

In this type of optical experiment the polarization of the sample is produced by absorption of a circularly polarized beam of resonance radiation propagating along the z axis.[17] The monitoring is carried out by the same beam as it passes through the sample and is detected. Since we have chosen a $J=1$ system as a representative example, our discussion would apply to the absorption of D_1 light by the $F=1$ (and by extension to the $F=2$) hyperfine levels of the $^2S_{\frac{1}{2}}$ ground state of Rb^{87} or Na^{23}, or the absorption of 10 830 Å light by the $^3S_1$ metastable state of He^4.[18]

The absorption of σ_+ light increases the angular momentum such that $\mu=m+1$, and in subsequent reemission little or none of this is lost. The system tends to polarization. Figure 3 shows an ideal case in which the transition is from a $J=1$ state to a $J=1$ state. This type of pumping can often be achieved by optical filtering of hyperfine components. Atoms which absorb will eventually find themselves in the state which does not absorb, so the population tends to be inversely proportional to the absorption rate. The system tends toward an ideal polarization[19] given by

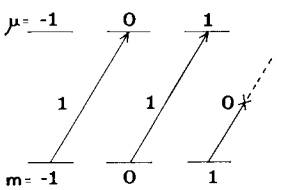

FIG. 3.

$$\varrho^0=\begin{vmatrix}1&0&0\\0&0&0\\0&0&0\end{vmatrix}. \tag{42}$$

The monitoring operator in this case is rather simple to determine. Figure 3 shows there are no interfering transitions, and so there are no off diagonal elements. From the matrix elements (41) we find the relative absorption rates from (40) shown on Fig. 3, and find that the absorption matrix is

$$\mathbf{A}=K\begin{vmatrix}0&0&0\\0&1&0\\0&0&1\end{vmatrix}.$$

Strictly speaking, we measure the absorption by means of the residual transmission of the beam, so that $\mathbf{M}=\mathbf{I}-\mathbf{A}$ or

$$\mathbf{M}=K\begin{vmatrix}1&0&0\\0&0&0\\0&0&0\end{vmatrix}, \tag{43}$$

[15] Reference 6, p. 167 contains a particularly brief discussion, or see Ref. 4 or 9.

[16] It sometimes happens that transitions are excited to several groups of levels in which the fine structure or hyperfine structure is not resolved. If the reduced matrix elements P_0 and P_1, for example, are of the same size, the optical components may nearly or completely cancel through the fact that the Clebsch–Gordon coefficients in (41) differ in sign for the cases $\Delta J=0$ and $\Delta J=1$, and the optical monitoring signal may be small or vanish. This does not affect the form of $\mathbf{M}$, but may mean that K vanishes.

[17] H. G. Dehmelt, Phys. Rev. **105**, 1487 and 1924 (1957).

[18] F. D. Colegrove and P. A. Franken, Phys. Rev. **119**, 680 (1960). See, also, T. R. Carver, Science **141**, 599 (1963).

[19] W. Franzen and A. G. Emslie, Phys. Rev. **108**, 1453 (1957).

which is similar to the state selector operator of the atomic beam. The resulting signal is

$$S=K(1-A-F/2). \tag{44}$$

E. The Brossel–Bitter Experiment

In a typical experiment[20] of this type the $6\,^1S_0$ ground state of mercury vapor (considered for simplicity to be an isotope with no nuclear spin) absorbs the $\sigma_+(\mathbf{e}=\hat{x}+i\hat{y})$ polarized resonance line at 2537 Å and makes a transition to the $6\,^3P_1$ $m_J=1$ state. Magnetic resonance occurs in the m levels of this excited state. The emitted fluorescence light acts as the monitoring light. If a linear analyzer oriented parallel to the z axis ($\mathbf{e}=\hat{z}$) is used, only the π component will be detected in the emitted radiation.

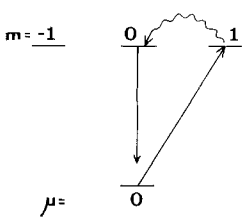

FIG. 4.

An examination of Fig. 4 shows that π polarized radiation is emitted from the $m=0$ state (to the $\mu=0$ state) only, so we find that

$$\mathbf{M}=K\begin{vmatrix}0&0&0\\0&1&0\\0&0&0\end{vmatrix}, \tag{45}$$

and the resulting signal is

$$S=KA. \tag{46}$$

From the standpoint of this discussion, the Brossel–Bitter experiment is a version of the Dehmelt experiment in which ideal polarization may be achieved, and both experiments have a monitoring operator similar to those of the atomic beam experiments. The function A, or Brossel–Bitter function, is a double peaked function when b is large, and is quite different from the ordinary Lorentz shape.

F. The Bell–Bloom Experiment

The polarization of the sample in this experiment[21] is produced in the same manner as in the Dehmelt experiment. The observation of the signal is with a cross beam of light perpendicular to the z axis, propagated in the y direction, say, and circularly polarized so that the polarization vector $\mathbf{e}$ is given by $\hat{x}+i\hat{z}$. With respect to the z axis of quantization, this light consists of σ_+, σ_- and π polarizations, and there are diagonal elements (d) of $\mathbf{M}$. However, in this experiment the photodetector is tuned only to the frequency ω. Only the nearest off-diagonal elements are important in giving an ω signal when taking the trace of $\varrho\mathbf{M}$. The effect of the farthest off-diagonal elements (f) is discussed later. Using Fig. 5 and the matrix elements (41) we find the form of $\mathbf{M}$ to be

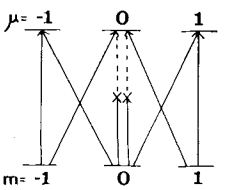

FIG. 5.

$$\mathbf{M}=K\begin{vmatrix}(d)&1&(f)\\1&(d)&1\\(f)&1&(d)\end{vmatrix}. \tag{47}$$

Drawing attention only to the nearest off-diagonal terms we can immediately see that the ω frequency Bell–Bloom signal has exactly the same form as that of the induction experiments (37), since the relevant part of the $\mathbf{M}$ is the same as (35). Indeed the classical models of the two experiments are the same; in the Bell–Bloom experiment the amount of scattered light changes as the particles precess from a parallel to an antiparallel direction in the cross beam.

G. Dodd–Series–Taylor Experiments[22]

For closest analogy to the preceding experiments we consider either a polarized ground state as in the Dehmelt experiment, or a polarized excited state as in the Brossel–Bitter experiment. Observation of the signal is carried out either by absorption of a transverse y axis beam which is polarized at 45° to the z axis, or by detection of emitted light which is examined by a similarly oriented analyzer. In either case we assume that the photodetector is sensitive only to the ω frequency. The only difference in the monitoring operator when compared to the Bell–Bloom type of experiment is that the light polarization is $\mathbf{e}=\hat{x}+\hat{z}$ instead of $\mathbf{e}=\hat{x}+i\hat{z}$. Yet the result when

[20] J. Brossel and F. Bitter, Phys. Rev. **86**, 308 (1952).
[21] W. E. Bell and A. L. Bloom, Phys. Rev. **107**, 1559 (1957). See also, H. G. Dehmelt, Phys. Rev. **105**, 1924 (1957).
[22] J. N. Dodd, G. W. Series, and M. J. Taylor, Proc. Roy. Soc. (London) **A273**, 41 (1963).

the **M** matrix is written out from (41) is that

$$M_{1,0}=P_0(+1)(\hat{x}+\hat{z})\cdot\hat{z}P_0 2^{-\frac{1}{2}}(\hat{x}+\hat{z})^*\cdot(\hat{x}+i\hat{y})$$
$$=2^{-\frac{1}{2}}(P_0)^2=K,$$
$$M_{0,-1}=P_0 2^{-\frac{1}{2}}(\hat{x}+\hat{z})\cdot(\hat{x}-i\hat{y})P_0(-1) \tag{48}$$
$$\times(\hat{x}+\hat{z})^*\cdot\hat{z}=-2^{-\frac{1}{2}}(P_0)^2=-K,$$

leading to **M** of the form

$$\mathbf{M}=K\begin{vmatrix}(d) & 1 & (f)\\ 1 & (d) & -1\\ (f) & -1 & (d)\end{vmatrix}. \tag{48'}$$

The resulting signal modulated at the frequency ω is

$$S=K[(X-X')\cos\omega t+(Y-Y')\sin\omega t]$$

or using (29)

$$S=K(B\cos\omega t+C\sin\omega t). \tag{49}$$

It cannot be emphasized too strongly that although there appear to be rather trivial differences between the Bell–Bloom experiment and the Dodd–Series–Taylor experiment, the lack of a simple classical model for the latter type of modulation experiment stimulated a comprehensive theory of light modulation effects.[5] One can see that either a "classical" Bloch resonance signal or a "Majorana" signal may be observed by altering the type of polarizer.[23] The reader may easily verify from the corresponding **M** matrix (dimension 2×2) for the spin $\frac{1}{2}$ system and (28), that although there is a Bell–Bloom signal from the spin $\frac{1}{2}$ system, there is no Dodd–Series–Taylor signal. The optical monitoring operator can distinguish between the two spins. (Of course, there are many other ways to determine the spin of a particle.)

H. 2ω Modulation

There are many aspects to the Dodd–Series–Taylor experiments. It will by now be obvious that the effect of the extreme off-diagonal elements (f) of the **M** matrix is to select the elements of the density matrix which have a time dependence at the frequency 2ω. All that is required is to choose monitoring light which connects the two states labeled by $m=\pm1$ only, as indicated in Fig. 6. This can be done by using a linear analyzer or polarizer which selects σ light, either linearly polarized and propagated in the z direction or polarized perpendicular to the z axis in the cross beam, since either is a coherent mixture of σ_+ and σ_- light with $\mathbf{e}=\hat{x}$ or $\hat{y}$. Since the two (f) elements are real and have the same sign, the resulting signal is

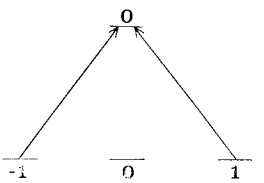

FIG. 6.

$$S=K(D\cos2\omega t+E\sin2\omega t). \tag{50}$$

It is worth pointing out here that the use of unpolarized light propagated along the z axis, which is described by a random mixture of σ_+ and σ_- or $\mathbf{e}=\hat{x}$ *and* $\hat{y}$, will not complete a coherent interference cycle in the sense of Fig. 2, and in fact, there are no modulation effects because the random mixture gives zero off-diagonal elements of M. It is also worth noting that the ability to see 2ω modulation components in the state of the system is a feature of the optical monitoring method which is not possible in induction resonance experiments. It is possible to think of the 2ω modulation as an effect of degenerate double-quantum absorption.[3]

I. Alignment Experiments

The Brossel–Bitter and Dodd–Series–Taylor experiments were originally carried out with π polarized light incident on mercury vapor, so that the sample was initially aligned rather than polarized. The initial density matrix corresponding to (20) for an *aligned* spin 1 system is

$$\varrho^0=\begin{vmatrix}1&0&0\\0&0&0\\0&0&1\end{vmatrix}\quad\text{or}\quad\varrho^0=\begin{vmatrix}0&0&0\\0&1&0\\0&0&0\end{vmatrix}. \tag{51}$$

The former may be represented by a superposition of $\rho_+{}^0$ and $\rho_-{}^0$, and the latter may then be formed by subtraction from the unit matrix **I**. The density matrix $\varrho(t)$ corresponding to these forms of ϱ^0 may be found (by inspection) by applying the same operations to expression (29). The reader may easily verify that the results are in agreement with the results of the Dodd–

[23] It has been pointed out to the authors by G. W. Series that this distinction may also be made in terms of selection rules on the quantum numbers in the rotating coordinate system of p. See Appendix.

Series–Taylor experiments, and also that there is no Bell–Bloom or induction signal in the case of alignment. It is well known in the case of induction experiments that one must have a polarized sample in order to see resonance signals.

J. Higher Order Experiments

It is beyond both the scope of this discussion and the power of the formalism developed so far to describe a large number of "carry-over," "indirect," or "second-order" phenomena, varieties of which are still being observed.[24] In these experiments, a first cycle of optical pumping produces initial coherence between the m levels under consideration, which is represented by off diagonal elements in ϱ^0. What is required in order to consider this type of problem in detail is a more complete picture of the initial excitation process[1,5] than we have given. We can give one example, however. Excitation of the $m=0$, $6\,^1S_0$ state of mercury to the $m_J=\pm 1$ states of the $6\,^3P_1$ state, if carried out with a coherent superposition of σ_+ and σ_- light represented by the inverse of Fig. 6, produces coherence between those states represented by extreme off diagonal elements labeled by $m_0=\pm 1$, $m_0'=\mp 1$. What is meant by coherence in this case is that the two states may be excited together by one Fourier component or mode of the light, if the two states are in a "level-crossing" situation[25] in which the Zeeman splitting between the states is small compared to the inverse lifetime, or width, of the states. The two states will not have arbitrary relative phase, and the products of their wavefunctions will not average to zero. This results in the off diagonal entries of ϱ^0.

Using the initial matrix

$$\varrho^0=\begin{vmatrix}1 & 0 & 1\\ 0 & 0 & 0\\ 1 & 0 & 1\end{vmatrix} \tag{52}$$

and (22) to work out the appropriate $\varrho(t)$, it can be seen that the terms

$$\exp[-i(m-m'-m_0+m_0')\omega t]$$

lead to 4ω frequency terms in the extreme off diagonal elements of $\varrho(t)$, and these when picked out by the (f) elements described by Sec. H lead to 4ω light modulation.[26] It is almost, but not completely, obvious that the form of the signal is that of the F function, since this is the function which in (29) is opposite the initially populated element of $\varrho_+{}^0$.

V. SUMMARY

By considering a density matrix for magnetic dipole resonance for the case of initial polarization alone it is possible to make a simple, unified and fruitful comparison of a large number of resonance experiments. In addition, the formalism developed here makes a convenient introduction to the theory of all the resonance line-shape functions. Each of the experiments may be described in terms of an initial preparation of states represented by ϱ^0, the development of the state represented by $\varrho(t)$, and the measurement represented by $\mathbf{M}$, in a familiar quantum mechanical manner. A description in this form makes it easier to see the similarities and differences between these experiments. We have not considered experimental details, or the magnitude of the signals involved, since these matters are simply and well described in the original literature.

One of the important conclusions to be noted is that whereas the atomic beam-monitoring method can measure only the diagonal or single state elements of the density matrix, and the induction experiments can measure only the nearest off diagonal elements in a restricted "classical" way, the optical monitoring operator can measure all of the $\Delta m=0$, ± 1, ± 2 elements with considerable flexibility of phase and, in particular, can measure all of the properties of the $J=1$ system. This added flexibility has its origin in the fact that we use a combination of two electric-dipole transitions, the selection rule for each of which alone is $\Delta m=0$, ± 1.

We conclude with an interesting and as yet incompletely answered question: what kinds of experiments and what sorts of corresponding monitoring operators would be capable of "measuring" the elements of a density matrix of par-

[24] O. Nedelec, M. N. Deschizeaux, and J. C. Pebay-Peyroula, Compt. Rend., **257**, 3730 (1963); K. Rosinski, Bull. Acad. Polonais Sci. Math., Ast. Phys. **12**, 497 (1964); R. B. Partridge and G. W. Series, to be published; B. P. Kibble and S. Pancharatnam, to be published.

[25] P A. Franken, Phys. Rev. **121**, 508 (1961).

[26] B. P. Kibble and G. W. Series, Proc. Roy. Soc. (London) **A274**, 213 (1963).

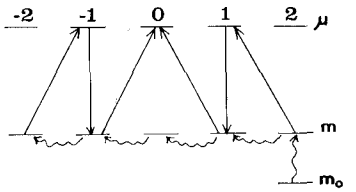

FIG. 7.

ticles with $J>1$? The question is particularly relevant to determining the off-diagonal elements, since it is obvious that the atomic beam-monitoring operator can measure the diagonal elements, no matter how numerous.

ACKNOWLEDGMENT

The authors wish to thank G. W. Series for many useful comments and discussions, and for his kindness and hospitality during their stay in England.

APPENDIX

Concerning the question raised at the end of this discussion, it is useful and important to note that the optical monitoring operator involving two electric dipole-matrix elements is limited in its ability to distinguish between particles with $J>1$ or to give new resonance functions. Just as the trace $(\varrho \mathrm{J}_x)$ does not contain resonance functions with terms in the denominator of form $\Gamma^2+(\Delta n)^2p^2$ with $\Delta n>1$, no matter what the value of J, so one would also expect that no such terms with $\Delta n>2$ would appear in the trace $(\varrho\mathbf{M})$ where $\mathbf{M}$ is the optical monitoring operator. This is equivalent to the previously mentioned selection rule, $\Delta n\leqslant 2$, which can be proved by transforming the $\mathbf{e}$ vectors corresponding to the Bell–Bloom or Dodd–Series–Taylor experiments to the rotating coordinate system aligned along the p axis and then examining the matrix elements $\langle n|\mathbf{e}'\cdot\mathbf{P}'|n'\rangle$ in that system.

One would therefore suppose that for all systems with $J>1$ the optical monitoring operators corresponding to the Bell–Bloom or Dodd–Series–Taylor experiments would give the same resonance functions as we have already derived for $J=1$. This may be shown for $J=\frac{3}{2}$, for example, by using the optical monitoring operators

$$\mathbf{M}=K\begin{vmatrix} 6 & \sqrt{3} & -\sqrt{3} & 0 \\ \sqrt{3} & 4 & 2 & -\sqrt{3} \\ -\sqrt{3} & 2 & 4 & \sqrt{3} \\ 0 & -\sqrt{3} & \sqrt{3} & 6 \end{vmatrix} \tag{A1}$$

and

$$\mathbf{M}=K\begin{vmatrix} 6 & 2\sqrt{3} & \sqrt{3} & 0 \\ 2\sqrt{3} & 4 & 0 & \sqrt{3} \\ \sqrt{3} & 0 & 4 & -2\sqrt{3} \\ 0 & \sqrt{3} & -2\sqrt{3} & 6 \end{vmatrix} \tag{A2}$$

for $\mathbf{e}=\hat{x}+i\hat{z}$ and $\mathbf{e}=\hat{x}+\hat{z}$, respectively, which describe the transitions from the m levels of a $J=\frac{3}{2}$ state to the μ levels of a $J=\frac{3}{2}$ state. Since the evaluation of a complete resonance density matrix for $\frac{3}{2}$ is tedious, it is simpler to show this by considering the numerators of the $\Gamma\pm i(\Delta n)p$ terms in combinations such as

$$\sqrt{3}\rho_{\frac{3}{2},\frac{1}{2}}+2\rho_{\frac{1}{2},-\frac{1}{2}}+\sqrt{3}\rho_{-\frac{1}{2},-\frac{3}{2}}, \quad \rho_{\frac{3}{2},\frac{1}{2}}-\rho_{-\frac{1}{2},-\frac{3}{2}} \tag{A3}$$

and

$$\rho_{\frac{3}{2},-\frac{1}{2}}+\rho_{\frac{1}{2},-\frac{3}{2}},$$

using (22) and the rotation operators for $J=\frac{3}{2}$. These three terms represent the relevant parts of the trace over the $\mathbf{M}$ operator for the Bell–Bloom or induction experiment, the Dodd–Series–Taylor ω modulation experiment or the Dodd–Series–Taylor 2ω modulation experiment, respectively. It should again be noted, just as for the $J=1$ case, that the nearest off-diagonal elements of the first of these operators (A1), which corresponds to the Bell–Bloom operator, are the same as those for the J_x operator for $J=\frac{3}{2}$.

In view of these limitations, it would appear that in order to construct monitoring operators which pick out elements of the density matrix with Δm or $\Delta n>2$ it would be necessary to make use of other than electric dipole transitions (very difficult in practice), or to use "carry over" experiments of the type illustrated by Fig. 7 (limited to "level-crossing" situations).