V. NUCLEAR MAGNETIC RESONANCE AND HYPERFINE STRUCTURE

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A. ATOMIC-BEAM LIGHT SOURCES

Studies of nuclear structure based on the nuclear perturbation of atomic energy levels may be divided into two classes; one class employs microwave techniques to measure the hyperfine structures directly, the other uses optical techniques in which the desired structures are inferred from differences in radiated optical frequencies. This report is concerned with possible improvements in the optical techniques which give promise of becoming an important supplement to the currently popular microwave-absorption experiments.

In conventional optical experiments, the detail of the hyperfine structure is limited both by the Doppler broadening of the radiated line and the resolving power of the apparatus. This note concerns the first problem: the reduction of the Doppler broadening of spectral lines radiated from a vapor. It should be pointed out that even if it proves difficult to push the resolving power of interferometers as far as might be desirable for the analysis of some very sharp lines, nevertheless the magnetic-scanning techniques developed in the M. I. T. Magnet Laboratory can be used to achieve a resolving power which is limited only by the width of the lines that are radiated or absorbed by a beam of atoms.

An atomic-beam light source is placed in a magnetic field, and the light which it radiates is then passed through an atomic beam absorber. The value of the scanning field for which the radiated frequencies are absorbed by the normal unperturbed atom can be used to calculate the frequency differences not only between the hyperfine components of a single atom, but also between lines radiated by various isotopes. An apparatus for this purpose is being built and will be reported on later. For certain lines, effective resolving powers of the order of 10^8, or more, seem possible.

In Fig. V-1 the Doppler width of lines radiated at room temperature by atoms with mass numbers in the range of 25 to 200 are plotted as a function of frequency or wavelength. It will be seen that resolving powers in the vicinity of 10^6 are required to match the Doppler widths. The ratio of the Doppler widths to the classical widths of allowed electric dipole radiation is indicated by the straight lines and the scale on the right. It will be seen that the Doppler widths range from 15 to 150 times the natural widths, and therefore that resolving powers from 10^7 to 10^9 would be required to extract all the information about nuclear structures contained in these lines. For some forbidden lines, even greater resolving powers would be needed. Since even this procedure will not give the detail that can be obtained on ground states with microwave

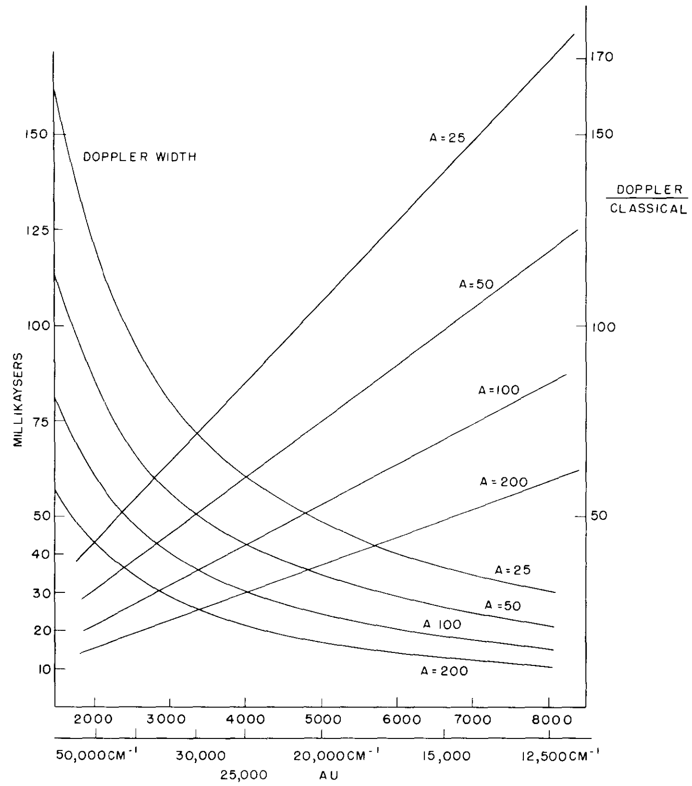

Fig. V-1. Chart showing Doppler and classical natural linewidths of spectral lines radiated from atoms of mass number A at room temperature.

measurements, it seems important to extend the observations to other energy levels, particularly to the study of isotope shift and nuclear-charge distribution.

While the use of atomic beams to reduce Doppler width is an old story (1), there seems to be no thorough discussion of the main drawback of atomic-beam light sources, that is , of their low intensity. To increase the light intensity coming from a given surface area in a specified solid angle, we may either increase the depth to which radiating atoms are present (up to the point where self-absorption of the center of the line broadens it) or increase the density of atoms in a small depth. Both of these methods are useful for different spectral lines and different optical instruments.

A system of beams for a deep source and for an absorption cell is shown in Fig. V-2 and Fig. V-3. In the lamp shown in Fig. V-2 a heated mercury container is thermally insulated from the refrigerated walls of a system of flat narrow passages. Quartz lamps illuminate the beam emerging from the slits. The absorbed and re-emitted resonance radiation is observed perpendicular to the aperture at A. A 1/200 degree of collimation is attempted. The construction of absorber and emitter are similar, in that thin flat mica spacers 0.001 inch by 1 inch by 1 inch are held apart by stainless

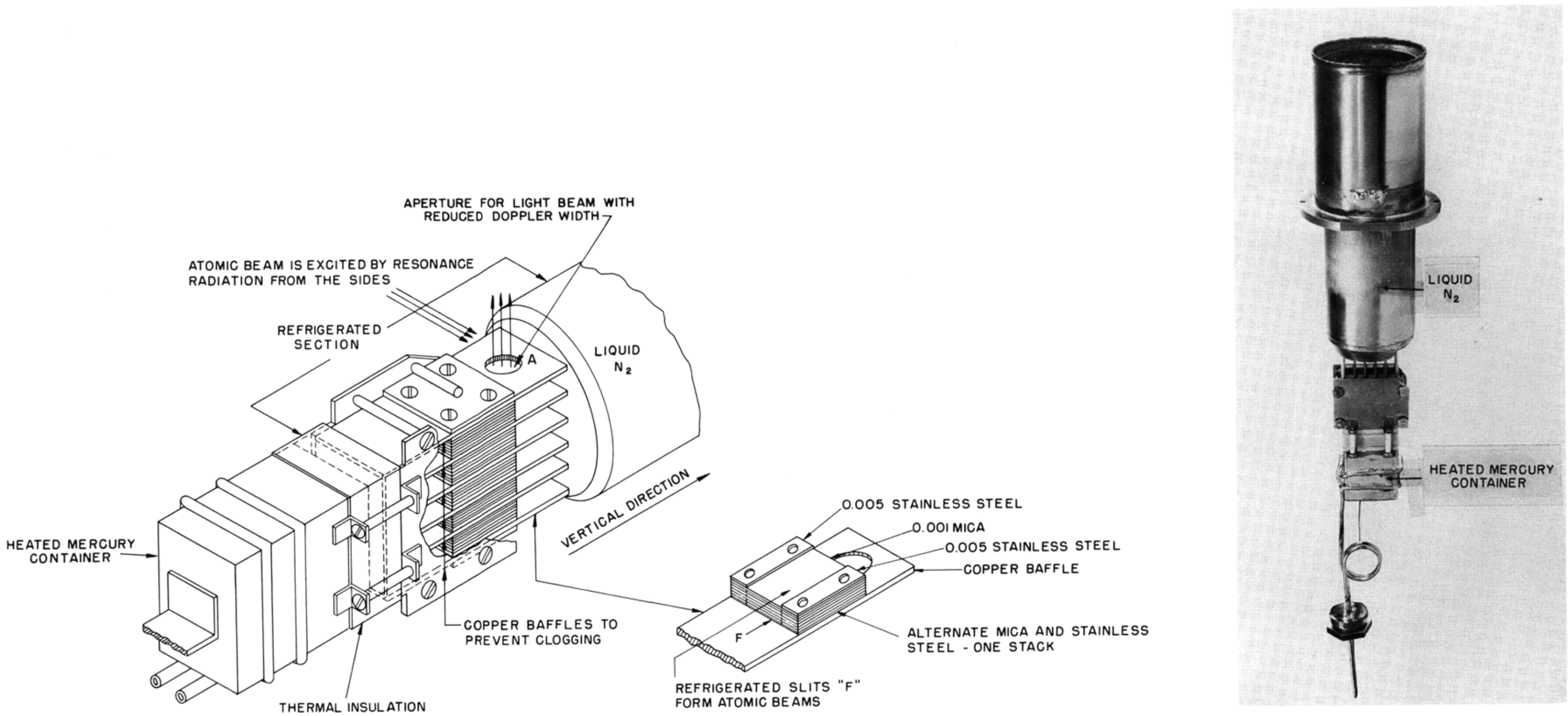

Fig. V-2. Schematic diagram and photograph of atomic-beam resonance lamp.

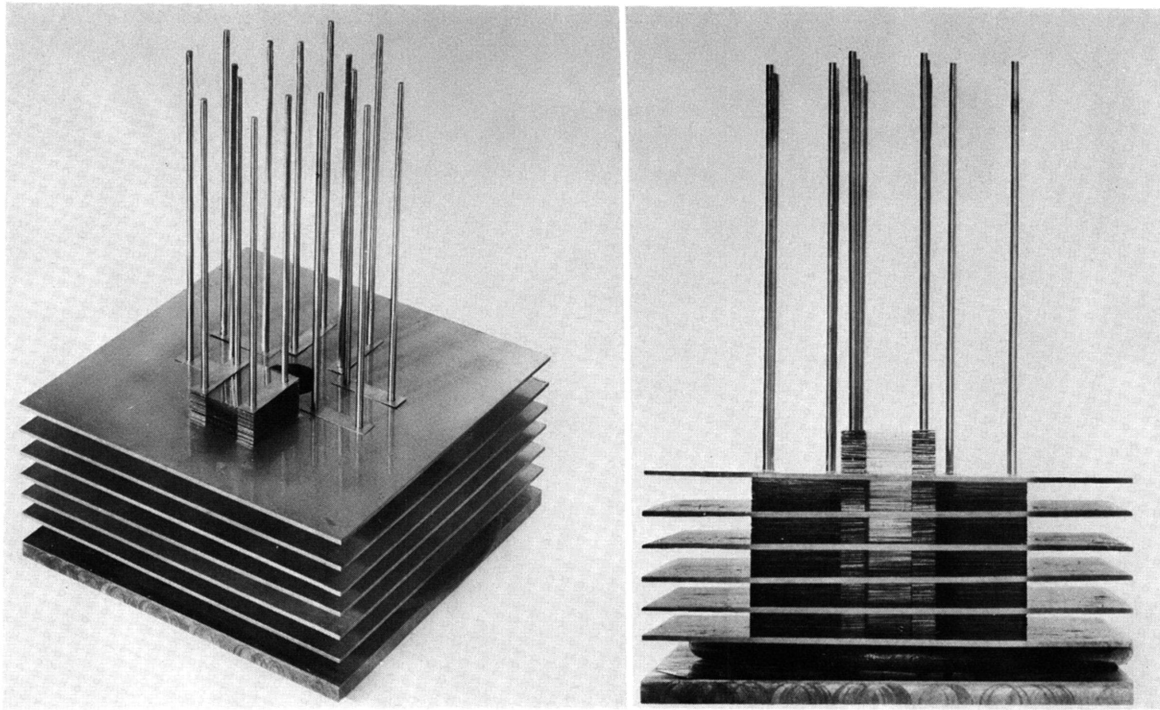

Fig. V-3. Two views of the atomic-beam absorption cell, partly assembled. Four intersecting beams are used. The light to be absorbed passes axially through the central hole.

steel pieces 0.005 inch by 0.25 inch by 1 inch. Baffles sop up most of the transversely moving mercury before it reaches the narrow collimating slots. It is hoped to attain densities in the useful portions of the beams of the order of 10^9 and to keep the apparatus in operation for many hours.

In applications which do not require a great depth of beam it is necessary to raise the atomic density to as high a value as possible. A simple argument, given below, leads to a useful conclusion. The maximum density of atoms in a beam at a distance r from an oven hole of radius $R_o \ll r$ is proportional to the angle of collimation $\phi = R_o/r$, and inversely proportional to r.

$$N_r = \frac{\phi}{8\pi\sigma^2 r} \tag{1}$$

Here $\pi\sigma^2$ is the atomic collision cross section. The density of atoms in the oven should be such that the mean free path of atoms in the oven is comparable to R_o. This expression is only intended as an order-of-magnitude guide. For $\phi = 1/50$, $\sigma = 3 \times 10^{-8}$ cm, r = 10 cm, we find that $n_r = 10^{11}/\text{cm}^3$, a number slightly smaller than the vapor pressure of mercury at −25°C. To prevent clogging in a single beam of the kind that is envisioned, a construction like that shown in Fig. V-4 may be used.

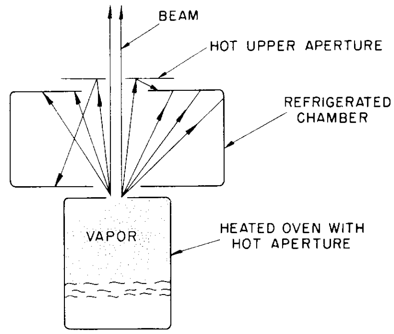

Fig. V-4. Proposed design for prevention of clogging of apertures in high-density beams.

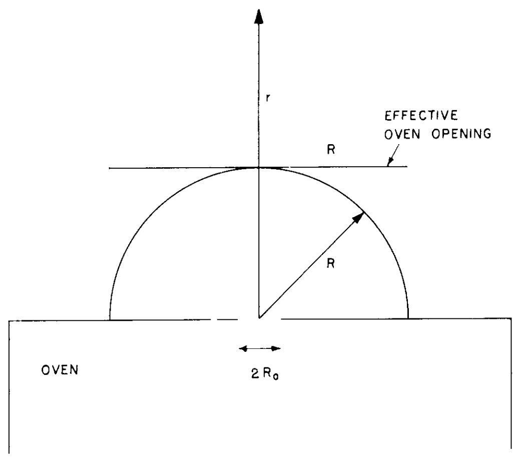

Fig. V-5. Definition of symbols: R is that distance from the oven opening at which $L_R = 10R$; n_o is the number of atoms/cc in the oven; L_o is the mean free path of atoms in the oven.

The argument leading to Eq. 1 is as follows. For a sufficiently small hole in an oven, the number of atoms escaping per second is simply the number that would have struck the area of the hole when it was closed.

$$N_o = n_o \bar{v} \pi R_o^2/4 \text{ atoms/sec} \tag{2}$$

n_o = number of atoms/cc in the oven

$\bar{v}$ = mean velocity of atoms in the oven

R_o = radius of the circular hole in the oven

Knudsen (2) has shown experimentally that Eq. 2 is satisfied for the range of pressures in which the mean free path in the oven L_o is greater than $10R_o$. As the oven pressure is increased, the rate of efflux increases by a factor of 2 over that predicted in Eq. 2 as L_o approaches R_o. Finally, as the pressure is further increased to values of $L_o = R_o/10$, the rate of efflux is still approximately twice that predicted by Eq. 2. We neglect this small variation, and assume that Eq. 2 is applicable to the ranges of oven pressures and apertures that are of interest.

In the range of oven pressures required for high-intensity beams, we shall assume $L_o \leqslant R_o$, and we define R as the distance from the aperture at which collisions may be neglected, which is the point at which $L_R = 10R$. (See Fig. V-5.) The approximate expressions for L_R and L_o are

$$L_R = \frac{1}{n_R \pi\sigma^2} = 10R \tag{3}$$

$$L_o = \frac{1}{n_o \pi\sigma^2} \tag{4}$$

Assuming that for r = R we already have a transport velocity of the order of the mean velocity in the oven, and a more or less uniform distribution over a hemisphere, we may set

$$N_o = n_o \bar{v}\, \pi R_o^2/4 = n_r\, 2\pi r^2$$

$$n_r = \frac{n_o}{8}\left(\frac{R_o}{r}\right)^2 \tag{5}$$

and, therefore, from Eq. 3 and Eq. 4, we obtain

$$\frac{L_R}{L_o} = \frac{10R}{L_o} = \frac{n_o}{R_R} = 8\left(\frac{R}{R_o}\right)^2$$

$$R = R_o \cdot \frac{R_o}{L_o} \tag{6}$$

In order to check the validity of our assumptions (the assumption for r > R collisions may be neglected), approximate calculations of the rate at which atoms leave a cylinder of radius R and length r − R through the end face, and through the sides as the result of collisions, were undertaken, These calculations indicate that if factors of the order of 2 are not important, then the value for R given in Eq. 6 may be adopted.

The result follows immediately if we define an effective angle of collimation ϕ' as $R/(r-R)$, or, since $R \ll r$, as $\phi' = R/r$. We have, from Eq. 5 and Eq. 6,

$$n_r = \frac{n_o}{8}\left(\frac{R_o}{r}\right)^2 = \frac{1}{L_o 8\pi\sigma^2}\left(\frac{R_o}{r}\right)^2$$

$$= \frac{1}{8\pi\sigma^2}\,\frac{R}{R_o^2}\left(\frac{R_o}{r}\right)^2$$

$$= \frac{\phi'}{8\pi\sigma^2 r} \qquad (7)$$

For maximum beam intensity, r should be small, and, therefore, the pressure should be low enough so that $R \approx R_o \approx L_o$. Equation 7 then reduces to the desired expression (Eq. 1).

An apparatus is being built to test this expression. It is hoped to extend the investigation presently to include the shaped nozzles suggested by Kantrowitz and Grey (3) and tested by Kistiakowsky and Slichter (4).

F. Bitter

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B. MULTIPLE QUANTUM TRANSITIONS IN MAGNETIC-MOMENT INTERACTIONS

The following report is a summary of a paper that is being prepared for submission for publication.

Transitions between two neighboring Zeeman levels induced upon a system by a steady magnetic field are commonly produced with an additional rf field of a frequency equal to this separation. Such transitions, however, have also been observed when the radio frequency equals one-half, one-third, one-fourth, and so on, of this same energy separation, and explanations for such observations have been made (1, 2, 3).

Among other things, our own investigations show the following: All of these resonances are deduced from classical theory, which gives a very good approximation to the complete time-dependent behavior of a Larmor system in a general rf field of one frequency. The experimental effect of the higher-order resonances may be as large as that of the first-order resonance. Moreover, once the classical behavior of the system is known, the quantum mechanical result can be written immediately, and be easily evaluated.

1. Classical Formulation of the Problem

Consider a Larmor system, which is governed by the familiar equations of motion:

$$\frac{d}{dt}\vec{J} = \gamma\,\vec{J} \times \vec{\mathscr{H}} = \vec{J} \times \vec{H}$$

where $\vec{J}$ is the total angular momentum; $\vec{\mathscr{H}}$ is the total magnetic field, which may be time-dependent; γ is the gyromagnetic ratio; and $\vec{H} = \gamma\vec{\mathscr{H}}$.

It is convenient to employ the well-known procedure (4) of viewing $\vec{J}$ in a rotating coordinate system specified by $\vec{\omega}(t)$. The equations of motion become

$$\frac{[d\vec{J}]}{dt}\,\vec{\omega} = \vec{J} \times (\vec{H} + \vec{\omega}) = \vec{J} \times \vec{H}_e$$

where $\frac{[d\vec{J}]}{dt}\,\vec{\omega}$ is the rate of change of $\vec{J}$ relative to the rotating system, and $\vec{H}_e$ (the effective magnetic field in the rotating frame) $= \vec{H} + \vec{\omega}$.

2. Classical Solution of Particular Problems

a. Stationary magnetic field: $\vec{H} = H_o\vec{k}$, $\frac{d}{dt}H_o = 0$

For $\vec{\omega} = -H_o\vec{k}$, $\vec{J}$ is at rest, or $\vec{J}$ precesses counterclockwise about the z-axis viewed from positive z to negative z. This motion may be written symbolically in terms of a classical rotation operator, $R_z(\theta)$, which, acting on a space vector, rotates that space vector about the z-axis by an angle, θ. Thus,

$$\vec{J}(t) = R_z(-H_o t)\,\vec{J}(0)$$

b. Stationary magnetic field along the z-axis and rotating field in the xy-plane: $H = H_o\vec{k} + R_z(-\omega' t)\,H_2\,\vec{i}$ (See ref. 4.)

By setting $\vec{\omega} = -\omega'\,\vec{k}$, or, equivalently, by applying $R_z(\omega' t)$ to all vectors, we find

$$H_e = (H_o - \omega')\,\vec{k} + H_2\,\vec{i}$$

Since the effective field is now stationary, the results of section a can be applied directly, to give

$$\vec{J}(t) = R_z(-\omega' t)\,R_{\vec{H}_e}(-H' t)\,\vec{J}(0)$$

where

$$H' = \left[(H_o - \omega')^2 + H_2^2\right]^{1/2}$$

$$\tan\theta = \frac{H_2}{H_o - \omega'}$$

θ = angle between $\vec{H}_e$ and $\vec{k}$.

In other words, there is precession of $\vec{J}$ about $\vec{H}_e$ and precession of $\vec{H}_e$ about the z-axis. When $\omega' = H_o$, J_z changes rapidly to give a resonance. The time, T, for a complete cycle in J_z is

$$T \doteq \frac{2\pi}{H_2}$$

c. Stationary field plus sinusoidal field along the z-axis and rotating field in the xy-plane: $\vec{H} = H_o\vec{k} - H_1\cos(\omega't)\,\vec{k} + R_z(-\omega't)\,H_2\,\vec{i}$

By suitable rotation about the z-axis, the term $-H_1\cos(\omega't)\vec{k}$ can be eliminated from the effective magnetic field. In the new frame, the field in the xy-plane rotates with an angular velocity that is not uniform. This field can be decomposed by Fourier analysis into an infinite sum of fields, each rotating with a different angular velocity that is always an integral multiple of ω'. Thus there will appear fields rotating at the frequencies ω', $2\omega'$, $3\omega'$, $4\omega'$, and so forth. Each field can produce a resonance if its frequency of rotation is near the Larmor frequency, H_o. Thus there will be resonances for $\omega' = H_o$, $2\omega' = H_o$, $3\omega' = H_o$, and so forth.

First let $\vec{\omega} = H_1\cos(\omega't)\vec{k}$, or, equivalently, apply to all vectors $R_z(\alpha)$, where

$$\alpha = \frac{-H_1}{\omega'}\sin(\omega't)$$

Then

$$\vec{H}_e = H_o\vec{k} + R_z(\alpha)\,R_z(-\omega't)\,H_2\,\vec{i}$$

$$= H_o\vec{k} + \sum_{n=-\infty}^{\infty} D_n R_z(n\omega't)\,H_2\,\vec{i}$$

It can be shown that

$$D_n \doteq \left(\frac{H_1}{2\omega'}\right)^{(n+1)} \frac{(-1)^{(n+1)}}{(n+1)!} \qquad n \geqslant -1$$

$$D_{-n} \doteq \left(\frac{H_1}{2\omega'}\right)^{(n-1)} \frac{1}{(n-1)!} \qquad n \geq +1$$

Section b shows that the effect of a rotating field is appreciable if and only if the frequency of that rotation is near the Larmor frequency, H_o. Thus, if we now assume that $n\omega' \doteq H_o$, then $\vec{H}_e$ can be written approximately as

$$\vec{H}_e \doteq H_o \vec{k} + R_z(-n\omega' t)\, D_{-n} H_2\, \vec{i}$$

This magnetic field has the same form as that with which section b begins, with $\omega' t$ replaced by $n\omega' t$, and H_2 replaced by $D_{-n}H_2$. Thus the results of that section can be taken over directly, if the effect of the rotation $R_z(\alpha)$ is included. Hence,

$$\vec{J}(t) \doteq R_z(-n\omega' t)\, R_z(-\alpha)\, R_{\vec{H}_{ee}}(-H_n' t)\, \vec{J}(0)$$

where

$$H_n' = \left[(H_o - n\omega')^2 + (D_{-n}H_2)^2\right]^{1/2}$$

$$\tan\theta_n = D_{-n} \frac{H_2}{H_o - \omega'}$$

$$\theta_n = \text{angle between } \vec{H}_{ee} \text{ and } \vec{k}$$

As before, the time for a complete cycle in J_z near a resonance of order n is

$$T_n \doteq \frac{2\pi}{D_{-n}H_2}$$

Thus for this special case, the classical theory predicts resonances for $n\omega' = H_o$, where n is any positive integer.

Many magnetic-resonance experiments measure the extent to which magnetic-moment orientation is destroyed. Such destruction occurs in the first-order resonance when T_o becomes smaller than the relaxation time of the system. Larger values of H_2 do not increase the height of the resonance. Accordingly, if T_n becomes smaller than the relaxation time of the system, the first n resonances should appear with equal heights.

d. General case: steady field and arbitrary rf fields of one frequency

The general case exhibits multiple resonances classically, but it will not be discussed here.

3. Quantum Mechanical Formulation of the Problem

Once a classical solution is obtained, as in the examples above, the quantum mechanical solution can be written immediately, and yields the same behavior as the classical.

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