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THE PERTURBING EFFECT OF TWO ROTATING FIELDS ON ANGULAR CORRELATIONS*

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ABSTRACT

The theory of nuclear magnetic resonance in perturbed angular correlations is extended to include the effects of two rotating magnetic fields, and their superposition to give an oscillatory field. If both radiations are parallel to the static field, the counter-rotating field gives a second order shift of the resonance (Bloch-Siegert effect). For other directions, there is a first order change in the line shape which depends on the azimuthal orientation of the counters.

I. INTRODUCTION

The time honored technique of magnetic resonance has recently found new applications to excited states of radioactive nuclei.[1] If the usual combination of static and periodic fields are applied, the resonance can be detected through a change in the angular distribution of the decay products. The theoretical line shapes of resonances produced by a single rotating field have already been analyzed.[2] We will consider the line shapes caused by a superposition of two rotating fields, and especially by a single oscillatory field.[3]

Although some of the customary formulas still apply, the resonances observed in an angular distribution are qualitatively different from the usual magnetic resonances. In the latter case, the line shape is given by the transition probabilities $|(m_2|\Lambda|m_1)|^2$, while the former depends as well on interference terms among the transition amplitudes. In the standard notation of angular correlation theory, the directional distribution is expressed in terms of the transition amplitudes through the parameters $G_{\ell_1\ell_2}^{q_1q_2}$, defined as

$$G_{\ell_1\ell_2}^{q_1q_2} \equiv \sqrt{(2\ell_1+1)(2\ell_2+1)} \sum (-)^{2I-m_1-m_2} \langle m_2|\Lambda|m_1\rangle\langle m_2'|\Lambda|m_1'\rangle^* \begin{pmatrix} I & I & \ell_1 \\ m_1' & -m_1 & q_1 \end{pmatrix} \begin{pmatrix} I & I & \ell_2 \\ m_2' & -m_2 & q_2 \end{pmatrix} \quad . \qquad (1)$$

By inverting this definition we can relate the transition probabilities to these parameters,

$$|\langle m_2|\Lambda|m_1\rangle|^2 = (-)^{2I-m_1-m_2} \sum \sqrt{(2\ell_1+1)(2\ell_2+1)}\ G^{0\,0}_{\ell_1\ell_2} \begin{pmatrix} I & I & \ell_1 \\ m_1 & -m_1 & 0 \end{pmatrix} \begin{pmatrix} I & I & \ell_2 \\ m_2 & -m_2 & 0 \end{pmatrix} . \quad (2)$$

This shows that the parameters $G^{0\,0}_{\ell_1\ell_2}$ alone suffice to determine the transition probabilities (as well as the angular distribution for both counters parallel to the axis). The idea of a resonance in $G^{q_1q_2}_{\ell_1\ell_2}$ is a generalization of a resonance in the transition probability, and the basic formulas must be rederived.

To do so, we must solve the Schrödinger equation[4]

$$i \frac{\partial}{\partial t} \psi(t) = - \underset{\sim}{I} \cdot \underset{\sim}{h}(t)\, \psi(t) \quad (3)$$

in the form[5]

$$\psi(t_2) = \Lambda(t_2,t_1)\, \psi(t_1) \quad (4)$$

and insert Λ into Eq. (1) to evaluate the time dependent parameters $G(t_2,t_1)$. For resonance experiments we are interested in the parameters $\overline{G}$

$$\overline{G}(t_1,\tau) \equiv \frac{1}{\tau}\int_0^\infty dt\ e^{-t/\tau}\ G(t_1+t,t_1) \quad (5)$$

averaged over the mean lifetime τ of the excited state. The $\bar{G}$ depend on the frequency and phase of the applied rf fields. We can take a further average over the phase $\Delta = \omega t_1$,

$$\bar{\bar{G}}(\tau) \equiv \frac{1}{2\pi}\int d\Delta \; \bar{G}(t_1,\tau) \quad . \tag{6}$$

When we refer to a line shape we will always understand the frequency to be constant, and the magnetic field to vary.

This program can be carried out <u>exactly</u> for a single rotating field [2]

$$h(t) = (h_1\cos\omega t, \; -h_1\sin\omega t \; , \; h_0) \tag{7}$$

using rotating coordinates. The transformation Λ is a sequence of rotations, and the parameters G can be expressed in terms of the matrices d^{ℓ} which represent these rotations. We want to use instead the magnetic field

$$h(t) = h_1(\cos\omega t, \; -\sin\omega t, \; 0) + h_2(\cos\omega t, \; \sin\omega t, \; 0) + h_0(0,0,1) \tag{8}$$

with strengths $h_0 \gg h_1,h_2$. There is no loss in generality in choosing the two fields to rotate in opposite directions at the same frequency ω, since two fields rotating at different frequencies ω_1, ω_2 can be brought to this form by choosing a frame rotating at frequency $(\omega_1+\omega_2)/2$. It saves a lot of complication to begin in this frame.

We will not attempt an exact solution of the problem. It leads to coupled differential equations similar to the Mathieu equation. The difficulty in finding an exact solution can be understood physically by recognizing that while the single rotating field Eq. (7) gives only one resonance (at $h_0 = \omega$), the double rotating field Eq. (8) gives more than just two resonances (at $h_0 = \omega, -\omega$); it also gives a transfinite number of multiple quantum resonances. What we want is not a line shape theory for all of these, but only the corrections to the fundamental resonance at $h_0 \simeq \omega$. The principal difficulty in deriving this approximation is to include the effects of "secular perturbations" which grow with time like $h_1 t$ and to discard only terms like (h_1/h_0). We will show that this can be done, to sufficient accuracy, by a sequence of rotations.[6] The results do not depend explicitly on the spin I, but only on the magnetic moment. More systematic methods would be necessary to extend our results to higher accuracy.[7]

II. MAIN DERIVATION

Consider transforming the Hamiltonian $H = -\underset{\sim}{I}\cdot\underset{\sim}{h}(t)$ by a sequence of rotations. Each rotation changes the direction of $\underset{\sim}{I}$, but leaves the form of the Hamiltonian unchanged. We can specify the Hamiltonian conveniently by giving the effective magnetic field in each frame. We begin with the field of Eq. (8), which clearly has terms which resonate at $h_0 \simeq \omega$. Our problem is to find a sequence of transformations $\psi \longrightarrow \psi_1, \psi_2, \psi_3 \cdots$ and $h \longrightarrow h_1, h_2, h_3 \cdots$, which finally remove such secular terms.

The first idea is to transform the field h_1 to rest, and the field h_2 to frequency 2ω. This succeeds in removing resonant terms, leaving only a small off-resonant field. If we were to neglect this field we would obtain the usual "zeroth order" approximation which predicts a single resonance at $h_0 = \omega$. A more accurate solution, which includes all of the first order effects of h_2 (and some of the second order), is given by the following transformation.

First transform the counter-rotating field to rest,

$$\psi_1 = e^{+i\omega t I_z}\,\psi$$
$$\underline{h}_1(t) = h_1(\cos 2\omega t, \; -\sin 2\omega t, 0) + (h_2, 0, h_0 + \omega) \quad . \tag{9}$$

Next rotate the static part of this field into the z-direction

$$\psi_2 = e^{i\theta I_y}\,\psi_1$$
$$\underline{h}_2(t) = h_1(\cos\theta \, \cos 2\omega t, \; -\sin 2\omega t, \; \sin\theta \, \cos 2\omega t) + (0,0,1)\sqrt{(h_0+\omega)^2 + h_2^2} \quad . \tag{10}$$

The angle θ is defined by

$$\tan \theta \equiv \frac{h_2}{h_0 + \omega} \tag{11}$$

and remains small throughout the resonance region. The effective field in this frame still has a resonance, but at a slightly displaced value of h_0.

To eliminate the resonant contributions, we once again rotate about the new z-direction at frequency 2ω,

$$\psi_3 = e^{-2i\omega t I_z}\,\psi_2$$

$$\underset{\sim}{h}_3(t) = h_1(-\sin^2\theta/2\ \cos 4\omega t,\ -\sin^2\theta/2\ \sin 4\omega t,\ \sin\theta\ \cos 2\omega t)$$
$$+\ (h_1\ \cos^2\theta/2,\ 0,\ \sqrt{(h_0+\omega)^2+h_2^2}\ -2\omega) \qquad (12)$$

and again bring the static part into the z-direction

$$\psi_4 = e^{i\beta I_y}\,\psi_3$$

$$\underset{\sim}{h}_4(t) = h_1(-\sin^2\theta/2\ \cos\beta\ \cos 4\omega t\ -\sin\theta\ \sin\beta\ \cos 2\omega t,\ -\sin^2\theta/2\ \sin 4\omega t,$$
$$-\sin^2\theta/2\ \sin\beta\ \cos 4\omega t + \sin\theta\ \cos\beta\ \cos 2\omega t)$$
$$+\ (0,0,\Omega)\ . \qquad (13)$$

The static field is now

$$\Omega = \left\{\left[\sqrt{(h_0+\omega)^2+h_2^2}\ -2\omega\right]^2 + h_1^2\ \cos^4\theta/2\right\}^{1/2} \qquad (14)$$

which is of order h_1 near the resonance region. The angle is defined by

$$\tan\beta = \frac{h_1\ \cos^2\theta/2\ \cos\theta}{h_0\ -\ \omega(2\cos\theta\ -1)} \qquad (15)$$

which passes rapidly through $\pi/2$ near the resonance. The field $\underset{\sim}{h}_4$ contains only small fields of order $h_1(\frac{h_2}{h_0})$ and $h_1(\frac{h_2}{h_0})^2$ at frequencies 2ω and 4ω, which are far off resonance. We will obtain a useful approximation by neglecting the effect of these time dependent fields, keeping only the static field Ω. This keeps the full dependence on h_1 and all the first order terms in h_2.

The perturbation matrix in the frame 4 is approximately

$$\Lambda_4(t_2,t_1) \cong e^{+i\Omega(t_2-t_1)I_z} \tag{16}$$

or transforming back to the original frame,

$$\Lambda \cong e^{-i\omega t_2 I_z} e^{-i\theta I_y} e^{+2i\omega t_2 I_z} e^{-i\beta I_y} e^{+i\Omega(t_2-t_1)I_z} e^{+i\beta I_y} e^{-2i\omega t_1 I_z} e^{+i\theta I_y} e^{+i\omega t_1 I_z} \quad . \tag{17}$$

Since this involves only pure rotations, it leads to a relatively simple expression for G. Making repeated use of the transformation of a tensor under rotations

$$D(R)\, T_\ell^q\, D^+(R) = \sum_p T_\ell^p \mathcal{D}^\ell_{pq}(R) \tag{18}$$

the transformation Eq. (7) gives

$$G_{\ell_1\ell_2}^{q_1q_2}(t_2,t_1) \cong \delta_{\ell_1\ell_2}\, e^{i\omega(q_1t_1 - q_2t_2)} \sum_{p_1p_2} d^{\ell}_{p_1q_1}(\theta)\, d^{\ell}_{p_2q_2}(\theta)\, e^{-2i\omega(p_1t_1 - p_2t_2)}$$

$$\times \sum_r d^{\ell}_{rp_1}(\beta)\, d^{\ell}_{rp_2}(\beta)\, e^{-ir\Omega(t_1 - t_2)} \quad . \qquad (19)$$

For the time averaged parameters $\bar{G}$ we find

$$\bar{G}_{\ell_1\ell_2}^{q_1q_2}(\Delta,\tau) \cong \delta_{\ell_1\ell_2}\, e^{i(q_1 - q_2)\Delta} \sum_{p_1p_2} d^{\ell}_{p_1q_1}(\theta)\, d^{\ell}_{p_2q_2}(\theta)\, e^{-2i\Delta(p_1 - p_2)}$$

$$\sum_r \frac{d^{\ell}_{rp_1}(\beta)\, d^{\ell}_{rp_2}(\beta)}{1 + i(q_2 - 2p_2)\omega\tau - ir\Omega\tau} \quad . \qquad (20)$$

Averaging the rf phase Δ reduces the number of summations by one

$$\bar{\bar{G}}_{\ell_1\ell_2}^{q_1q_2}(\tau) \cong \delta_{\ell_1\ell_2} \sum_{p_1p_2} d^{\ell}_{p_1q_1}(\theta)\, d^{\ell}_{p_2q_2}(\theta)\, \delta_{q_1 - q_2,\, 2p_1 - 2p_2}$$

$$\sum_r \frac{d^{\ell}_{rp_1}(\beta)\, d^{\ell}_{rp_2}(\beta)}{1 + i(q_2 - 2p_2)\omega\tau - ir\Omega\tau} \quad . \qquad (21)$$

This can be further simplified by passing to the limit $\tau \longrightarrow \infty$

$$\lim_{\tau\to\infty} \bar{\bar{G}}_{\ell_1\ell_2}^{q_1q_2} \cong \delta_{\ell_1\ell_2} \; d^{\ell}_{p_1q_1}(\theta) \; d^{\ell}_{p_2q_2}(\theta) \; d^{\ell}_{0p_1}(\beta) \; d^{\ell}_{0p_2}(\beta) \tag{22}$$

if $q_{1,2} = 2p_{1,2}$ are even, and zero otherwise.

The three Eqs. (20)-(22) give our main results, expressed in terms of d^{ℓ} functions and the parameters θ, β, Ω. It is easily seen that as $h_2 \longrightarrow 0$, these formulas approach the exact results for a rotating field. For h_2 finite, they give a useful approximation to the parameters G in the vicinity of $h_o \simeq \omega$. We will not try to show the results in graphical form because the changes are quite small, but will only try to discuss the results analytically.

III. DISCUSSION

1. In order to recognize some familiar results, consider first the coefficient $G_{\ell\ell}^{00}$. From Eq. (22) we find

$$\lim_{\tau \to\infty} \bar{\bar{G}}_{\ell\ell}^{00} = |P_{\ell}(\cos\theta)\; P_{\ell}(\cos\beta)|^2 \;. \tag{23}$$

The factor $P_{\ell}(\cos\theta)$ is nearly unity over the resonance line, with value

$$P_{\ell}(\cos\theta) \cong 1- \frac{\ell(\ell+1)}{16} \left(\frac{h_2^2}{h_0^2}\right) \tag{24}$$

and with slope of order $(h_1 \cdot h_2^2/h_0^3)$. The factor $P_\ell(\cos\beta)$ gives a ℓ-fold splitting of the resonance line, but at a field slightly shifted by the dependence of $\cos\beta$ on h_2. The line center is at $\cos\beta = 0$, or

$$h_0 = \omega(2\cos\theta - 1) \cong \omega - \frac{h_2^2}{4\omega} \quad . \tag{25}$$

To order (h_2^2/h_0^2) there is no change in the line shape, but only this shift. Our method is not sufficiently accurate to treat the higher order changes in shape, but taken literally Eq. (23) predicts a third order decrease in the separation of the minima and a third order asymmetry of the line.

The line shift in Eq. (25) is called the Block-Siegert shift, and is a well known effect. Our derivation shows that this is the dominant effect of the counter-rotating field for resonance experiments measuring the transition probabilities, and for angular correlation experiments with radiations parallel to h_0.

2. For other values of $q_1, q_2 \neq 0$, some of the parameters $\overline{G}$ have stronger dependence on h_2. We will consider only the results contained in Eq. (22). The dependence on h_2 is mainly contained in the small angle $\theta \simeq (h_2/2h_0)$. For $\theta = 0$ (rotating field approximation) only the terms $G_{\ell\ell}^{00}$ are non-zero. As we slowly increase the counter rotating field h_2, there are some terms which grow linearly with θ $(G_{\ell\ell}^{02}, G_{\ell\ell}^{20})$, others that grow quadratically $(G_{\ell\ell}^{22}, G_{\ell\ell}^{04}, G_{\ell\ell}^{40})$, etc. If only the first order terms are desired then we need only compute $G_{\ell\ell}^{00}$, $G_{\ell\ell}^{02}$, $G_{\ell\ell}^{20}$ and we can use

$$\tan\beta \cong \frac{h_1}{h_0 - \omega} \qquad \Omega \cong \left[(h_0 - \omega)^2 + h_1^2 \right]^{1/2} \quad .$$

The result will be to give a $\cos\ 2\phi$ azimuthal dependence of the line shape, proportional to h_2. In the rotating field approximation the line shape is independent of ϕ after averaging over the phase Δ, but the effect of the counter-rotating field is to give some dependence on the orientation of the counters relative to the plane of the rf field.

This effect can be understood in elementary terms by remembering that the angular distribution is sensitive to any change in direction of the spin, whereas the usual magnetic resonance can only observe changes in the energy (i.e. the z component of the spin). The first order terms are due to a rotation of the spin about the z axis through a small angle of order $\theta \sim (h_2/2h_0)$. We have not been able to think of any practical use of this effect, except perhaps as a means of directly measuring the strength of the rf field; this can also be done by observing the splitting of the peaks in $G^{00}_{\ell\ell}$ for the parallel configuration.

3. For a single rotating field, and in our approximation to two rotating fields, G has only diagonal terms $\ell_1 = \ell_2$. This can be traced to the perturbation matrix Λ being expressible as a sequence of pure rotations. This is an exact property of the single rotating field, but only approximately true for two rotating fields. Since we have emphasized the importance of "off-diagonal" terms $\ell_1 \neq \ell_2$ in testing symmetries,[8] it is of interest to discover just how small these terms are. We have shown that the off-diagonal terms are of order $\left(\frac{h_1^2 h_2^2}{h_0^4}\right)$, which is much too small to be of interest in PAC symmetry tests.

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REFERENCES AND FOOTNOTES

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4. For convenience, we choose $\hbar$ as the unit of action and μ as the unit of magnetic moment. This is equivalent to measuring magnetic fields in frequency untis (or vice versa).
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