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Eckart Matthias

January 19, 1967
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RECENT APPLICATIONS OF PERTURBED ANGULAR CORRELATIONS**†

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ABSTRACT

A brief review is given about the present-day experimental situation in the field. The applicability, limitations, and obtainable accuracy of the various existing methods are discussed, and the possibility of applying perturbed angular correlation and distribution techniques to isomeric levels with half lives longer than $10^{-6}$ sec is considered. The use of angular correlations or distributions to detect NMR in excited nuclear states is described in detail. Various source structures are examined with regard to their applicability in perturbed angular correlation work.

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I. INTRODUCTION

The purpose of this brief review is to sketch the recent experimental
development in the field of perturbed angular correlations (PAC). Considerable
progress has been made during the past three years, since the field was last
reviewed at the Uppsala meeting on Perturbed Angular Correlations. In parti-
cular, three types of experiments have had a great impact on the whole field
concerned with measurements of electromagnetic moments and hyperfine interactions
of short-lived nuclear levels:

1) Implantation of recoil atoms into ferromagnetic lattices following
Coulomb excitation, which, combined with a systematic knowledge of magnetic
hyperfine fields, will make it possible to measure g-factors for short-lived
first vibrational and second rotational states.

2) NMR in radioactive states with half lives down to $10^{-8}$ sec is possible
using angular correlations or distributions to detect the resonance. For short half
lives ($10^{-3}$ sec to $10^{-8}$ sec), the experiment has to be performed by making use
of some kind of intrinsic enhancement of the external rf field. For half lives
longer than $10^{-3}$ sec, the externally applied rf amplitude is sufficient to
destroy the angular distribution directly. The basic problem in all these
experiments, however, is that the polarization of the nuclei has to be preserved
for times longer than the lifetime of the state in order to obtain a detectable
destruction effect of the rf on the angular distribution.

3) The more general subject of perturbed angular distributions also
includes reorientation effects in Coulomb excitation which provides a means of
measuring quadrupole moments of excited nuclear states without relying on cal-
culated electric field gradients or nuclear models.
We will discuss here only the conventional perturbed angular correlation methods, and, in addition, the development of radioactive NMR detection. No attempt is made to cover the field thoroughly, and for a detailed and complete description of all conventional methods, the reader is referred to the articles by Steffen and Frauenfelder. 6

Perturbed angular correlation measurements with radioactive isotopes and perturbed angular distributions of gamma rays following Coulomb excitation have been widely used to determine magnetic moments of short-lived excited, nuclear states. 7 Systematic nuclear information has been gathered for $g_R$-factors of the first excited rotational states in deformed nuclei and for $g$-factors of first excited vibrational states in spherical nuclei. Although the PAC technique has been applied to the investigation of hyperfine fields, the information obtained in this way is to date far from systematic. In most of the experiments the approach was rather to determine the nuclear $g$-factor by using the known value for the magnetic hyperfine field, obtained from either Mössbauer effect results or nuclear magnetic resonance studies. Some systematic data on hyperfine fields for rare earth impurity atoms in iron have recently been measured by the recoil implantation technique following Coulomb excitation, 2 and the specific advantages of this method will undoubtedly trigger more of these types of investigations. Increased accuracy will bring a more detailed understanding of the field acting on the recoil nuclei and possibly indicate whether there is a unique field, several discrete values, or a field distribution.

Apart from a few exceptions, all the work with perturbed angular correlations or distributions has been carried out with short-lived states with half lives in the nanosecond or subnanosecond range, where the accuracy is
limited by the small rotation angles of the correlation pattern. It is recognized, on the other hand, that for half lives long enough to employ the time-differential method, good accuracy, of the order of one percent, can be obtained. The precision with which interaction frequencies can be measured improves with increasing half life. Surprisingly enough, however, there has not yet been one measurement on a state with a half life longer than 500 nsec. Up to now, no method for measuring magnetic moments has been proved to work for isomeric states with half lives between $10^{-6}$ sec and 1 sec. It seems feasible, however, that the PAC method could be applied at least up to the millisecond range. Although only a limited number of states with lifetimes in this range are known to be populated by cascading radioactive decay, many more isomeric levels can be conveniently produced by nuclear reactions. The generally non-isotropic angular distribution of gamma rays following the reaction process lends itself to either spin rotation or NMR experiments. The basic difficulty of all measurements involving long-lived levels will be to preserve the nuclear orientation for times longer than the half life.

The applicability of the method of perturbed angular correlations and distributions for different half-life ranges is illustrated in Fig. 1. The experiment of Sugimoto et al. proved that the polarization of the product nuclei in nuclear reactions can, under special circumstances, be preserved up to about 10 sec, which makes it possible to use the angular distribution as a detector for the NMR in the isomeric states. For the long-lived isomers, it is hoped that the range can be extended up to the half lives where nuclear orientation and atomic beam measurements become possible. Time differential spin-rotation measurements will dominate in the region $10^{-6}$ sec to $10^{-9}$ sec,
although some NMR experiments in that range might be possible. For half lives shorter than $10^{-9}$ sec, only time-integral measurements can be performed. For low-lying excited states in which the recoilless resonance absorption is possible, Mössbauer effect measurements are in direct competition with PAC in the range $10^{-10}$ sec to $10^{-6}$ sec. In practice, however, both techniques are often complementary in their application.

II. CONVENTIONAL PAC TECHNIQUES

A. Time-integral Method

If the resolving time, $2\tau_o$, of the fast-slow coincidence system is large compared to the mean life, $\tau$, of the intermediate nuclear state, $2\tau_o \gg \tau$, the measurement integrates over the exponential decay and one records what is called the time-integral angular correlation,

$$\hat{W}(\theta) = \frac{1}{\tau} \int_0^\infty e^{-t/\tau} W(\theta, t) dt$$

(1)

For a pure static magnetic dipole interaction with the magnetic field oriented perpendicular to the detector plane Eq. (1) has the form

$$\hat{W}_1(\theta, \gamma) = \sum_{k \text{ even}} \frac{b_k}{\sqrt{1 + (k\omega_L\gamma)^2}} \cos k(\theta - \Delta \theta)$$

(2)

with the abbreviations
\[ \Delta \theta = k^{-1} \arctg k \omega_L \tau , \quad 0 < \Delta \theta < \frac{\pi}{2k} \]

\[ b_2 = \frac{3/4 A_2 + 5/16 A_4}{1 + 1/4 A_2 + 9/64 A_4} \]

\[ b_4 = \frac{35/64 A_4}{1 + 1/4 A_2 + 9/64 A_4} \]

where \( A_2 \) and \( A_4 \) are the conventional angular correlation coefficients. In case there is a time-dependent perturbation of the form \( G_k(t) = e^{-\lambda_k t} \) superimposed on the magnetic dipole interaction, the integration leads to the expression

\[ \hat{W}_\perp(\theta, \Pi) = 1 + \frac{1}{4} A_2 \hat{G}_2 \left\{ 1 + \frac{3 \cos 2(\theta - \Delta \theta_{22})}{1 + (2\omega_L \tau \hat{G}_2)^2} \right\}^{1/2} \]

\[ + \frac{1}{64} A_4 \hat{G}_4 \left\{ 9 + \frac{20 \cos 2(\theta - \Delta \theta_{24})}{1 + (2\omega_L \tau \hat{G}_4)^2} \right\}^{1/2} + \frac{35 \cos 4(\theta - \Delta \theta_{44})}{1 + (4\omega_L \tau \hat{G}_4)^2} \]

(3)

where \( \Delta \theta_{nk} = n^{-1} \arctg n \omega_L \tau \hat{G}_k \), \( 0 < \Delta \theta_{nk} < \frac{\pi}{2n} \)

and \( \hat{G}_k = (1 + \lambda_k \tau)^{-1} \).

From Eqs. (2) and (3) one can see that the magnetic field causes two effects: an attenuation and a rotation of the angular correlation pattern. The rotation is only present if the detectors can distinguish between \( \gamma_1 \) and \( \gamma_2 \) of the cascade. Observable rotations are always smaller than \( 45^\circ \) for \( k=2 \), since for larger rotation
angles, the angular correlation is attenuated to isotropy. For very small rotations, the attenuation effect is negligible and \( \Delta \theta \approx \omega_L \tau \), i.e., the effect of the interaction on the angular correlation is linear. It is for these small values of \( \omega_L \tau \) where the time-integral angular correlation is a very valuable method, in particular, when used together with large internal fields. The magnitude of the hyperfine fields permits us to measure magnetic moments for nuclear levels with half lives down to \( 10^{-12} \) sec. An example demonstrating the accuracy which can be obtained for the quantity \( \omega_L \tau \) is shown in Figs. 2, 3 and 4. These measurements, done by the Hamburg group,\(^9\) give \( \omega_L \tau \) with an uncertainty of \( \pm 1.5\% \) for Ir\(^{193}\) (Fig. 2), \( \pm 6.7\% \) for Ru\(^{100}\) (Fig. 4), and \( \pm 1.7\% \) for Pd\(^{106}\) (Fig. 4). In spite of this accuracy for \( \omega_L \tau \), the final result for the product \( g \cdot H_{\text{eff}} \) is much smaller, as the nuclear mean life \( \tau \) is, in most cases, not known with a comparable accuracy. This characterizes the general situation for time-integral perturbed angular correlations involving mean lives in the pico-second region. The final result for either the nuclear g-factor or the effective field usually carries a rather large error arising from the uncertainty in the mean life \( \tau \).

The main application of time-integral measurements is the determination of magnetic dipole interactions for levels with mean lives between \( 10^{-12} \) and \( 10^{-9} \) sec. This can also be done without any polarizing external magnetic field. The integral PAC for such a randomly oriented magnetic interaction has the form\(^10\)

\[
\hat{W}(\theta) = \sum_{k \text{ even}} A_k \hat{g}_k P_k (\cos \theta) \quad (4)
\]
The attenuation factors are independent of the nuclear spin and given by

\[ \hat{G}_k = \frac{1}{2k+1} \sum_{N} \frac{1}{1+(N\omega_L\tau)^2} \]  

(5)

In the limit of a very strong interaction the attenuation factor approaches a limiting value, the "hard core", given by

\[ \hat{G}_k (\omega_L \tau \rightarrow \infty) = \frac{1}{2k+1} \]  

(6)

Thus for a large interaction strength, the measurement of \( \hat{G}_k \) does not lead to a unique value of \( \omega_L \tau \).

Two other possibilities are the determination of relaxation times and of static electric quadrupole interactions. A time-integral angular correlation perturbed by a relaxation interaction in the source is given by Eq. (4) with a perturbation factor

\[ \hat{G}_k = \frac{1}{1 + \lambda_k \tau} \]  

(7)

The relaxation parameter \( \lambda_k \) depends on the nature of the fluctuating interactions. Detailed expressions for electric and magnetic relaxations are given by Abragam and Pound.\(^8\) The important point to note is that the angular correlation can be attenuated to isotropy by a time-dependent relaxation in the source. However, to measure the time-integral angular correlation (Eq. (4)) is not a very sensitive way to obtain the relaxation parameter \( \lambda_k \), because it is integrated over the parameter of interest, the time.
Not many investigations of static electric quadrupole interactions have been carried out, probably because it is felt that the product $Q \cdot \frac{\partial \mathbf{E}}{\partial z}$, which can be determined from the measurements, is not of too much interest, unless one finds a way to calculate the field gradient $\frac{\partial \mathbf{E}}{\partial z}$. The formalism distinguishes two cases, that of a polycrystalline sample and that of a single crystal. The polycrystalline case corresponds to that of a random magnetic interaction and the angular dependence of the interaction Hamiltonian is averaged out. The attenuation factor, $\hat{G}_k$, in Eq. (4) is given in Ref. 8 for various spins $I$. Again, even for a very strong interaction, $\omega_E \tau$, the angular correlation is never completely wiped out and $\hat{G}_k$ approaches the "hard core value" which is given by Eq. (6) for half-integer spins and which is slightly larger for integer spins. The determination of the $\hat{G}_k(\omega_E \tau)$ is not a very sensitive method of obtaining $\omega_E \tau$ for polycrystalline sources, in particular, not for large values of $\omega_E \tau$. A non-axially symmetric quadrupole interaction influences the behavior of $\hat{G}_k(\omega_E \tau)$ slightly but it will be hard to detect this effect in practice.

In the case of a single-crystal source, we have as an additional parameter the angle between the major crystalline axis and the detectors which permits a good accuracy for the determination of $\omega_E \tau$. A few very attractive measurements have been carried out with single crystals, but their detailed description would go beyond the scope of this report and the reader is referred to the original references.
B. **Time-differential Method**

The time-differential method applies in all cases, where the electronic resolving time is small compared to the nuclear mean life, $\tau_0 \ll \tau$. Experimentally, it is of great importance since the time-dependent observation of the interaction matrix elements

$$\langle m_b | e^{-i/\hbar \hat{H}_t} | m_a \rangle \langle m_b' | e^{-i/\hbar \hat{H}_t} | m'_a \rangle^*$$

allows one to obtain the interaction frequency with high accuracy. It is worth noting that the time-spectrum of the perturbed angular correlation is the Fourier transform of the frequency spectrum obtained in NMR. Both techniques are related by the Fourier-transformation

$$f(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} g(\omega) e^{i\omega t} d\omega$$

$$g(\omega) = \int_{-\infty}^{+\infty} f(t) e^{-i\omega t} dt$$

In the limit of a large number of rotations of the angular correlation pattern, the accuracy approaches that usually obtained in NMR work. But even with a few observed cycles, the time-differential method is far superior to all time-integral measurements not only because of higher accuracy, but also because it is independent of the mean life $\tau$. Further, it shows directly any additional perturbation present in the source, either in form of amplitude damping or by the occurrence of several superimposed frequencies. A limitation imposed on the time-differential method is that the rotation of the angular correlation pattern
should be sufficiently slow that the electronic resolving time permits one to
distinguish between successive rotations, which means the condition \( \omega_L \tau_0 < \pi \)
has to be fulfilled. Very often this makes it impossible to measure magnetic
hyperfine fields in this way.

For the simplest case, a pure magnetic dipole interaction, oriented per-
pendicular to the detector plane, the coincidence rate between \( \gamma_1 \) and \( \gamma_2 \) detected
at an angle \( \theta_0 \) as a function of time can be written

\[
C(\theta_0, t)^\pm = e^{-t/\tau} \sum_{k \text{ even}} A_k P_k (\cos(\theta_0 \pm \omega_L t))
\]

where the \((+\) and \((-\) signs refer to opposite field directions. The exponential
decay is usually averaged out by forming a ratio, for example,

\[
R = 2 \frac{C(\theta_0, t)^+ - C(\theta_0, t)^-}{C(\theta_0, t)^+ + C(\theta_0, t)^-}
\]

Two typical examples of a time-differential PAC measurement are shown
in Figs. 5 and 6. From such data, the interaction frequency \( \omega_L \) can be obtained
with an accuracy of much better than one percent. This is often sufficient to
reveal fine details such as the Knight shift. It should be noticed that for a
time range as long as 1 \( \mu \)sec there is no attenuation of the angular correlation
pattern visible. This clearly demonstrates that cubic metal lattices are very
suitable as perturbation-free host lattices up to the microsecond range. The
usefulness of the time-differential technique for magnetic hyperfine fields
becomes obvious from Figs. 7 and 8. Measurements can also be performed with
randomly oriented magnetic fields as, for example, a completely demagnetized
ferromagnetic sample. In this particular case, the perturbation factor has the simple form

$$G_k(t) = \frac{1}{2k+1} \sum_{N} \cos N \omega_L t$$

(11)

which means that a superposition of k frequencies is observed. An example for this is shown in Fig. 9. Here, the frequency could be obtained with good accuracy from the time spectrum taken at just one angle. For small amplitudes, however, one has to measure the anisotropy as a function of time.

When trying to use the conventional time-differential method on half lives longer than 1 μsec, one runs into the following basic difficulties:

a) the source strength has to be reduced to keep the ratio of true to accidental coincidences acceptable; b) the time resolution of analog converters is limited and of the order 1% of the total time range; c) the limited time resolution requires the use of low magnetic fields. In addition, one has the usual problems with linearity, stability, and, in most instances, a memory limitation in the analyser. One experimental approach to overcome all the above weaknesses is a technique, called "Digital Analysis of Perturbed Angular Correlations" (DAPAC).13 The principle is to obtain the data directly in a digital form and to find the interaction frequency by calculating the Fourier transform, Eq. (8), of the time spectrum. The use of a digital time-to-height converter automatically offers the advantages that: 1) the resolving time is independent of the time range used; 2) there is practically no limitation in channel numbers; 3) it has perfect linearity and much greater stability than analog systems; and 4) the need for an accurate time calibration is completely avoided.
The data analysis is based on the fact that the Fourier transform of the time spectrum $W(t)$ is a function of frequency, $g(v)$, with the resonance frequency given by $v_L = \frac{2gH}{h}$. After normalization for the exponential decay the time spectrum can be described by the function $W(t) = \sum_k A_k P_k (\cos(\theta_L t))$ for the case of an oriented magnetic dipole interaction. The even and odd parts of the Fourier transform are calculated separately,

$$A(v) = \int_{T_{\min}}^{T_{\max}} W(t) \sin 2\pi vt \, dt, \quad B(v) = \int_{T_{\min}}^{T_{\max}} W(t) \cos 2\pi vt \, dt.$$  \hspace{1cm} (12)

The phase is defined as

$$\phi(v) = \tan^{-1} \left[ \frac{-A(v)}{B(v)} \right],$$

in accordance with the normal sign convention of the Fourier transform. This allows the determination of the sign of the magnetic dipole interaction for a certain detector geometry and field direction.

To illustrate this technique, two examples are shown in Figs. 10 and 11. The shift of the resonance in Fig. 11 is the Knight shift for Rh$^{100}$ in the three different metals. The most striking practical advantage of this method is that it is very time-saving. Since the analysis is based on a frequency search, it does not require such good statistical accuracy as is normally needed when a mathematical function is fitted to the data. Compared to the result of Figs. 10 and 11, it is possible to improve the signal-to-noise ratio more than an order of magnitude by using the autocorrelation function $C(T)/C(0)^{13}$ instead of $W(t)$ in Eq. (12). The autocorrelation technique smoothes the discrete data carrying a statistical uncertainty at each point.
Relaxation processes described by the attenuation factor

\[ G_k = e^{-\lambda_k t} \]  \hspace{1cm} (13)

can be investigated with the time-differential method. In quite a few cases the anisotropy has been observed as a function of time and the relaxation parameter \( \lambda_k \) has been determined, mostly for the purpose of obtaining the corrected nuclear g-factor (see Eq. (3)). There have also been attempts\(^{14}\) to experimentally verify a suggestion by Dillenburg and Maris\(^{15}\) that the attenuation factor in Eq. (13) does not always describe the relaxation interaction properly, and should, in some cases, be substituted by a sum of two exponentials. No final conclusion on this subject has been reached, but there is no convincing experimental evidence yet of such an effect and the theory of Abragam and Pound\(^8\) seems to describe all practical cases.

Time-dependent investigations of electric quadrupole interactions have also been carried out in some cases.\(^{16,12c,12d}\) These time spectra are very often difficult to interpret and one has to expect higher-order effects such as non-axial symmetry and frequency distributions.\(^{11}\) Again, a thorough discussion of this subject is beyond the scope of this paper.
III. RESONANCE DESTRUCTION OF ANGULAR DISTRIBUTIONS

A. NMR in Angular Correlations

As discussed above, one of the limitations of the time-differential method is that it cannot be applied if the time resolution is larger than the precession time for one cycle, \( \tau_0 \geq T_L = \pi / \omega_L \). This is often the case for magnetic hyperfine fields at impurity atoms in ferromagnetic lattices. We know, on the other hand, from Eq. (8), that the accuracy of a time-differential measurement improves as the number of observed spin rotations increases. This leads necessarily to the question: Is it possible to perform an NMR experiment with the excited state in a case where the rotations of the angular correlation pattern can no longer be resolved? The rf-field would then, at resonance, change the non-isotropic population of the substates (see Fig. (12)) and thus have a destructive effect on the angular correlation. This effect could then in turn be used for a radiative detection of NMR.

This question has been discussed as early as 1953 by Abragam and Pound, and the following conditions for such an experiment were formulated:

1. The splitting of the substates of the intermediate state (Fig. 12) should be large compared to the natural line width, i.e., \( \omega_0 \tau \gg 1 \).

2. The rf-field, \( H_\perp \), must have sufficient amplitude to induce transitions between the substates in times comparable to the nuclear lifetime, i.e., \( \omega_\perp \tau \gg 1 \), where \( \omega_\perp \) is the interaction frequency with a field of amplitude \( H_\perp \).

3. Nuclear relaxation times should be longer than the nuclear lifetime, \( \tau < \tau_{rel} \), so as not to broaden or wipe out the resonance line. Conditions (1) and (3) can easily be met, but (2) is so stringent that a resonance experiment was considered not to be feasible. For a lifetime of 0.2 \( \mu \)sec and a \( g \)-factor of one, for example, one
would have to use an rf amplitude of 1 Kgauss which would correspond to a power of $10^6$ Watt in $1 \text{ cm}^3$ at 40 MHz.

It is possible, however, to fulfill condition (2) by making use of the enhancement of the effective rf amplitude, $H_{\text{eff}}$, in nuclear magnetic resonance in ferromagnets. If the resonance is observed in a domain, the externally applied rf field, $H_\perp$, is amplified according to the relation

$$2 H_{\text{eff}} = \left(1 + \frac{H_{\text{hf}}}{H_0}\right) H_\perp$$

(14)

where $H_{\text{hf}}$ is the magnetic hyperfine field and $H_0$ is the external polarizing field. This means that the nuclear resonance is driven indirectly through the hyperfine coupling between the nucleus and the electrons, rather than directly by the external rf field. With the usual magnitude of magnetic hyperfine fields, it is easy to obtain an enhancement factor $H_{\text{hf}}/H_0$ of $10^3$ or larger which means that it is possible to meet condition (2) with external rf amplitudes of about 1 gauss.

The geometry for such a resonance experiment is shown in Fig. 13. A thin foil of the ferromagnetic material into which the activity has been diffused is mounted with its plane parallel to the axis of either one or both detectors. The polarizing field is applied parallel to this axis which ensures, for a sufficiently large $H_0$, a completely decoupled (unperturbed) angular correlation. The rf-field, $H_\perp$, has to be applied perpendicular to the detector plane and consequently, to $H_0$. The first resonance destruction effect detected with such an arrangement is shown in Fig. 14. For this experiment the $84-74.8$ keV cascade in Rh$^{100}$, which has a large positive anisotropy and a half life of
235 nsec, was used. The resonance was driven by the hyperfine field of Rh in Ni, which is 200 Kgauss. With the detectors arranged at 180°, one observes a decrease in the coincidence counting rate around the point of resonance. The natural line width due to the lifetime is about 0.5 MHz, which is much smaller than the observed one. The interpretation of the large line width is difficult, but newer results show that the resonance can actually be resolved into two components, corresponding to two different sites, a main peak and a satellite about 20 MHz lower. Whether there is an additional broadening due to relaxation phenomena cannot be determined until there is a theoretical prediction of the line width available.

Another example of a resonance with Rh$^{100}$ in Fe at two different temperatures is given in Fig. 15. The shift between room temperature and 77°K is obvious. The resonance is much more symmetric compared to the Ni case, but the observed line-width is still much larger than the natural line-width. Despite this, it is apparent that the interaction frequency $\omega_L$ can be determined from these data with an accuracy of about 0.1%.

The formalism to describe the resonance destruction has to be developed from the general expression for a PAC

$$W(t; k_1, k_2) = \sum_{k_1, k_2} A_{k_1} A_{k_2} B_{k_1 k_2}(t) \left[ (2k_1 + 1)(2k_2 + 1) \right]^{-1/2}$$

$$\times \psi_{k_1}^{\ast} (\theta_1, \phi_1) \psi_{k_2} (\theta_2, \phi_2)$$

(15)

where the perturbation factor is defined as
The interaction Hamiltonian is now time-dependent and has the form well-known from NMR theory:\(^{18,19}\)

\[ \mathcal{H}(t) = -\gamma \hbar \left[ H_0 I_z + H_1 (I_x \cos \omega t + I_y \sin \omega t) \right] \] (17)

The time dependence of \( \mathcal{H}(t) \) can be eliminated by transforming to a coordinate system which rotates about \( H_0 \) (z-axis) with frequency \( \omega \). With \( \vec{H}_1 \) along the x-axis we get a transformed (effective) Hamiltonian\(^{18,19}\)

\[ \mathcal{H}' = -\gamma \hbar \left[ \left( \frac{\omega}{\gamma} + H_0 \right) I_z + H_1 I_x \right] \] (18)

For the experimental set up described in Fig. 13, we have \( \theta_1 = 0^\circ \) and \( \theta_2 = 180^\circ \), which implies \( N_1 = N_2 = 0 \) and consequently, \( m_a = m_a' \) and \( m_b = m_b' \). With these simplifications, we find the time-integrated perturbation factor from Eq. (16).

\[
\alpha_{00}^{k_1 k_2} = \frac{1}{(2k_1 + 1)(2k_2 + 1)} \sum_{m_a m_b} (-1)^{2I + m_a + m_b} \begin{pmatrix} I & I & k_1 \\ m_a - m_0 & 0 \\ m_b - m_0 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m_a - m_0 & 0 \\ m_b - m_0 \end{pmatrix} \\
\times \frac{1}{\tau} \int_0^\infty e^{-t/\tau} \left| \langle m_b | e^{-i \hbar t} | m_a \rangle \right|^2 dt
\] (19)
The transformation into a rotating coordinate system effects only the interaction Hamiltonian, and not the angular correlation function, since the axis of rotation is chosen parallel to the emission direction of the \( \gamma \) rays. The square of the matrix element is the probability \( P_{m_{b}m_{a}}^{m_{b}}m_{a} \) of finding the spin \( I \) at time \( t \) in the state \( m_{b} \) when it was in the state \( m_{a} \) at time \( t = 0 \). The matrix elements are non-diagonal which means that the perturbation factor \( \mathcal{G}_{k_{1}k_{2}}^{00} \) is best calculated by numerical methods. It is interesting to note that the interference term for \( k_{1} \neq k_{2} \) in Eq. (19) is not zero. From Eqs. (15) and (19), it follows that the time-integrated angular correlation function exposed to a destructive rf field with the geometry shown in Fig. (13) is described by

\[
\mathcal{W}(180^\circ) = \sum_{k_{1}k_{2}} A_{k_{1}}(1) A_{k_{2}}(2) \cdot \mathcal{G}_{k_{1}k_{2}}^{00} \tag{20}
\]

with

\[
\mathcal{G}_{k_{1}k_{2}}^{00} = [(2k_{1} + 1)(2k_{2} + 1)]^{1/2} \sum_{m_{a}m_{b}} (-1)^{2I + m_{a} + m_{b}} \begin{pmatrix} I I k_{1} \\ m_{a} \end{pmatrix} \begin{pmatrix} I I k_{2} \\ m_{b} \end{pmatrix} \\
\times \langle n|m_{b}\rangle^{*} \langle n|m_{a}\rangle \langle n^{'i}|m_{b}\rangle \langle n^{'i}|m_{a}\rangle^{*} \frac{1}{1 + [(E_{n} - E_{n^{'i}})/\hbar]^{2}} \tag{21}
\]

In this equation \( E_{n} \) are the eigenvalues and \( \langle n|m \rangle \) the corresponding eigenvectors of the interaction Hamiltonian given in Eq. (18).
B. NMR in Statically Polarized Nuclei

The resonance destruction method to detect NMR in a radioactive state can always be employed when the nuclei in this state are oriented. In angular correlations the coincidence between $\gamma_1$ and $\gamma_2$ selects a certain non-isotropic population of the nuclei in the $m$-substates. Thus, the $\gamma_2$ are emitted from an oriented system with respect to $\gamma_1$. Another way of obtaining a polarization of the nuclei is to go to very low temperatures and expose them to a sufficiently large magnetic field, $H_{\text{eff}}$, so that $kT \ll H_{\text{eff}}$. This technique is well-known and discussed in detail in the literature. Other techniques for orienting nuclei at low temperatures will not be considered here.

The angular distribution of $\gamma$ rays emitted from polarized nuclei is given by

$$W(\theta) = \sum_{k \text{ even}} B_k U_k A_k P_k(\cos \theta)$$  \hspace{1cm} (22)$$

The coefficients $A_k$ are the same as $A_k^{(1)}$ or $A_k^{(2)}$ in the angular correlation function (see Eq. (15)). The coefficients $U_k$ account for any disorientation by preceding but unobserved transitions. The orientation parameter, $B_k$, describes the degree of nuclear orientation and is given by

$$B_k(I) = \sqrt{2I+1} \sum_m (-1)^{I-m} (I \ I \ m - m \ | k \ 0) \frac{e^{-E_m/kT}}{\sum_{m'} e^{-E_{m'}/kT}}$$  \hspace{1cm} (23)$$

where $(I \ I \ m - m \ | k \ 0)$ is a Clebsch-Gordon coefficient and $E_m$ are the eigenvalues of the interaction Hamiltonian. Resonance-induced transitions disturb the
Boltzmann distribution and, consequently, alter the orientation parameters $B_k$. The resonance effect should then be observable as a change in radiation intensity at a fixed angle $\theta$.

An experiment of this type was suggested, in principle, by Bloembergen and Temmer in 1953\textsuperscript{22} but no successful experiment has been carried out until very recently.\textsuperscript{23} The great difficulty is the large heating effect of the rf-field. However, this can be avoided by again taking advantage of the rf-amplitude enhancement in a ferromagnetic sample. According to Eq. (14), the external rf-field can be reduced by a factor of about $10^3$, leaving a still sufficiently large effective rf-amplitude acting at the nucleus. In this way, an adequate warm-up rate of the sample can be obtained. Note that there are no power requirements here due to a short nuclear lifetime. All radioactive levels suitable for nuclear polarization work have long enough lifetimes to eliminate this problem.

The geometry for such an experiment is sketched in Fig. 16. The radioactive material is diffused into a thin foil of the ferromagnetic host material. The foil is soldered on to copper fins which provide the thermal contact with the demagnetization salt. A set of superconducting Nb rings produce the polarizing field, $H_O$, oriented parallel to the plane of the foil. $\mathbf{R}_o$ defines the reference axis for the emission direction $\theta$ of the $\gamma$ rays. The rf-field is applied perpendicular to the polarizing field, but parallel to the plane of the foil. The effect of the rf on the angular distribution was first observed for Co\textsuperscript{60} in Fe.\textsuperscript{23} Figure 17 shows the warm-up rate for Co\textsuperscript{60} nuclei as a function of time and frequency (upper part) and with a fixed frequency off resonance (lower part). Summing up the results of repeated demagnetization runs lead to resonance
curves like the ones displayed in Fig. 18 which were obtained with two different alloys. Although the resonance frequency is the same, the two measurements give a different line width. We attribute the larger line width of alloy I to such experimental conditions as e.g., a larger inhomogeneity of the polarizing field. Alloy I was larger than alloy II and also two frequency scans were made following one demagnetization. The resonance for each second scan was shifted toward lower frequencies either because of a change of the trapped magnetic flux in the Nb rings or because of long relaxation times. The measurements with alloy II are more representative for a typical line width to be expected in these type of experiments. However, to draw any conclusions from the observed line width, one would have to use a more controlled external polarizing field with greater homogeneity.

Independent of these line width considerations, it is apparent that the interaction frequency can be obtained from these measurements with an accuracy of about 0.1%. This is at least an order of magnitude better compared to what can be done with conventional nuclear polarization experiments\(^{20,21}\) and underlines the importance of the method. Further, in contrast to conventional measurements, the rf destruction of the polarization gives the interaction frequency independent of any precise knowledge of the temperature scale. It should be noted that the conventional nuclear orientation technique measures an energy \(|\mu \cdot H_{\text{eff}}|\) while the present method gives a frequency \(|g H_{\text{eff}} \frac{\hbar}{\hbar}|\). Combining both techniques would determine the spin of the nuclear state. If a sufficiently large polarizing field is applied to completely saturate the ferromagnetic sample, the resonance is observed in domains. In that case, the resonance frequency should shift linearly with the applied field\(^{17}\) which would make it
possible to determine the nuclear $g$-factor directly. The technique will also prove valuable for studies of the magnetic hyperfine fields of impurities in ferromagnetic lattices. To a good approximation it measures the hyperfine field at 0°C and is capable of working with extremely small concentrations. A great number of radioactive isotopes and isomeric levels are accessible to this technique, particularly in connection with isotope separators, which provide a way to shoot isotopes into a lattice in cases where chemical procedures fail.

C. **NMR in Nuclei Polarized by Nuclear Reactions**

Nuclear reactions is another technique in which an appreciable degree of nuclear polarization is achieved. In particular, in (heavy-ion, xn) reactions, the product nucleus is highly aligned and the gamma rays emitted in the decay of the compound state show angular distributions with considerable anisotropies. If an isomeric state is populated in this decay, the high degree of alignment could be used to measure electromagnetic interactions in that state. One great advantage of this technique would be that very high spin states could be measured, as they are populated with a high yield in reactions with large momentum transfer. Also systematic measurements of the same spin state in different isotopes could be carried out. A practical advantage is that these measurements involve only singles counting, which eliminates restrictions in the available half-life range caused by accidental coincidences. However, the greatest difficulty which has to be overcome in such an experiment is to preserve the spin orientation in the isomeric level for a period longer than
the lifetime $\tau$. Here, the large recoil velocity of the product nuclei is very valuable in getting the nucleus quickly into a suitable environment.

Since the first experiment of Goldring and Scharenberg\textsuperscript{25} angular distributions following Coulomb excitation have been frequently used\textsuperscript{7} to measure nuclear $g$-factors by observing the effect of an external magnetic field on the distribution function. Very recently this technique has been extended to the use of internal magnetic fields\textsuperscript{2} by using the recoil to implant the Coulomb-excited nuclei into ferromagnetic lattices. However, due to the Coulomb excitation process this technique will be limited to nuclear levels in the nano- and subnano-second range. It will, therefore, hardly be possible to use it for resonance destruction experiments. The first perturbed angular distribution experiment involving a half life longer than $10^{-8}$ sec was performed by P. B. Treacy\textsuperscript{26} on the 198 keV level in $\text{F}^{19}$, excited in a $(p,p')$ reaction on $\text{F}^{19}$. This level has a half life of 87 $\mu$sec, very convenient for time-differential investigations. Some more work has been done with this level,\textsuperscript{27} but it still remains the only case where an angular distribution following a nuclear reaction was used for time-differential investigations involving a longer half life.

In a remarkable experiment Sugimoto et al.\textsuperscript{4} proved that the polarization of nuclei produced by nuclear charged particle reaction can be preserved up to 10 sec or longer at room temperature and subsequently used to detect the nuclear magnetic resonance. The principal idea for this experiment was to work with free ions and decouple the nuclear spin completely from the highly excited electronic states in a strong external field. To achieve this, Sugimoto et al.\textsuperscript{4} used the experimental set-up shown in Fig. 19. The recoil atoms were
taken out of the target, separated from the beam and caught by a CaF$_2$ recoil stopper. This crystal was chosen because it is cubic and known to have a long relaxation time for F nuclei. The reaction used was $^{16}\text{O}(d,n)^{17}\text{F}^{+}0^{+}$ with a half life of 66 sec for the $\beta^+$-decay of $^{17}\text{F}$. The polarization of the $^{17}\text{F}$ nuclei was detected by observing the asymmetry of the $\beta$-decay (see Fig. 19). The resonance effect is shown in Fig. 20.

This experiment sets a standard for what can be done with nuclear alignment following reactions. It demonstrates that, indeed, the whole half-life range from nanoseconds up to seconds is open for investigations. The decoupling technique used by Sugimoto et al. is probably the only way to avoid disorientation of the nuclei by hyperfine interactions with the highly excited ionic state, and would have to be used in the range between $10^{-6}$ sec and 1 sec. There is also evidence that thick cubic metallic targets are a suitable environment to observe unperturbed angular distributions up to the microsecond range. However, no measurements of electromagnetic interactions involving half lives longer than $10^{-8}$ sec have been made yet with metallic targets.

The polarization of the product nuclei following capture of polarized thermal neutrons should also be mentioned this section. D. Connor utilized this fact to determine the magnetic moment of $^{6}\text{Li}$. The NMR in this system was measured by using the asymmetry of the $\beta$-decay of polarized $^{6}\text{Li}$ nuclei for the detection. In contrast to charged particle reactions there is no change or disruption in the electronic shell involved for thermal neutron capture and hyperfine and relaxation interactions in the target are of the same nature as encountered in ordinary NMR.
IV. SOURCE STRUCTURE CONSIDERATIONS

Any attempt to use time-differential and resonance perturbed angular correlation techniques for half-lives longer than a microsecond faces the major difficulty that the source structure has to be such that time-dependent and static internal perturbations are small enough not to cause an appreciable disorientation during the time interval used for the measurement. There is no need to discuss internal perturbations of static character, as those only produce attenuation factors which vary periodically with time but never wipe out the angular correlation pattern completely.\(^6\) Our concern are time-dependent perturbations which damp the amplitude of the angular correlation pattern with time. Time-dependent interactions provide a relaxation mechanism through which the nuclei can lose their orientation. The originally non-isotropic population of the substates of the intermediate level becomes randomized as a result of transitions induced by the fluctuating fields. As the time behavior of the angular correlation pattern represents the Fourier transform of the NMR frequency spectrum, all facts about nuclear relaxation times in NMR spectroscopy apply in full to PAC measurements. The only, but important, difference is that here we have the lifetime of the intermediate state which sets a natural time window for the observation. Everything that happens outside this window cannot be observed with angular correlation techniques, i.e., small line broadenings are not measurable as the natural line width is comparatively large. Thus, the ratio of the nuclear lifetime, \(\tau\) to the relaxation time, \(\tau_{\text{rel}}\) is the factor which determines the proper choice of the source material. For very long relaxation times, \(\tau_{\text{rel}} \gg \tau\), the observed angular correlation pattern is undamped. If the relaxation time comparable to the lifetime, \(\tau_{\text{rel}} \sim \tau\), then
the angular correlation is strongly damped and an interaction frequency, \( \omega_L \), can only be measured if it is sufficiently large, \( \omega_L \tau_{\text{rel}} \gg 1 \). For still shorter relaxation times, \( \tau_{\text{rel}} \ll \tau \), the angular correlation is completely wiped out. In this discussion it was assumed that the coupling between the fluctuating fields and the nuclear moments is reasonable strong. The angular correlation remains, of course, unperturbed in all cases where the fields fluctuate so fast that the coupling is broken and the fields average to zero within a short time interval. This happens when \( \omega_{\text{rel}} \tau_c \ll 1 \), where \( \omega_{\text{rel}} \) is the relaxation interaction and \( \tau_c \) is the time interval during which a local field configuration remains stable.

In the following we discuss the three most commonly used source structures with regard to their applicability to longer half lives:

1. Liquid Sources and Ionic Crystals. Most g-factor measurements on nuclear states in the nanosecond range have been performed with liquid sources. From these measurements, one can extract the general result that nuclei in paramagnetic ions in liquids are affected by very strong time-dependent perturbations which destroy the angular correlation within a few nanoseconds at room temperature. In diamagnetic liquids, on the other hand, the anisotropy remains unperturbed for very long times. An example for a diamagnetic liquid source in which the anisotropy survived for more than 1.6 \( \mu \text{sec} \) was the measurement by Walter et al. on the 206 keV 0.57 \( \mu \text{sec} \) level of Re\(^{187}\). This is, in fact, up to now, the longest half life used for perturbed angular correlation measurements. From NMR measurements it is known that nuclei in diamagnetic liquids have extremely long relaxation times in the range of milliseconds to seconds or sometimes even longer. The difference between NMR measurements
and angular correlation experiments is, however, that in the latter the radioactive decay creates highly excited paramagnetic ions. The time in which the atom reaches its ionic ground state depends, to a large extent, on the atomic environment and varies from source to source. If the ionic ground state is diamagnetic and the atomic shell reaches it in a time short compared to the precession time of the nucleus in the external field (as, e.g., in metals), the angular correlation will not be further attenuated and the situation is similar to NMR measurements. If the ionic ground state is reached in times comparable or longer than the nuclear precession time, coherence is lost and the angular correlation is wiped out by this "after effect" perturbation. The existence of after effects in non-metallic sources is established by many experiments, but quantitative results are still lacking. The orientation of the nuclei can, however, be preserved by decoupling the nuclear and the electronic spin by a large enough external magnetic field. Such a decoupling experiment was done for after effects following α-decay and K-conversion.

A similar perturbation is present if the ionic ground state is paramagnetic. The nuclei will then precess in an effective field, $\mathbf{H}_{\text{eff}}$, which is given by the sum of the external $\mathbf{H}_0$, and the internal magnetic field, $\mathbf{H}_i$. It is usually described in terms of the paramagnetic correction factor, $\beta$.

$$\mathbf{H}_{\text{eff}} = \beta \mathbf{H}_0$$  (24)

Typical values for $\beta$ are tabulated for the tri-valent rare earth ions. Also, for ions of the 3d, 4d, and 5d elements, unless the element forms diamagnetic complexes, a substantial hyperfine structure may be present, modifying the
effect of the external magnetic field. A theoretical calculation of $\beta$ is not possible for those cases where the atom can exist in several oxidation states, since the distribution of atoms in these oxidation states is unknown after conversion processes, EC, and $\beta^-$ decay. If, however, the oxidation state and its hyperfine coupling constant is known, the correction factor $\beta$ can be calculated. The hfs constant can be measured by performing a decoupling experiment in the way described above. This was done by R. Stiening and M. Deutsch\textsuperscript{33} in the case of $^{156}\text{Ga}^{3+}$.

Another time-dependent perturbation very frequently found in liquid sources is caused by the interaction between the nuclear quadrupole moment and the fluctuating electric field gradients.\textsuperscript{8} Sources which show such an effect should be avoided for $g$-factor measurements instead of going into the trouble to measure the relaxation constant (see Eq. (13)).

2. Non-magnetic Metallic Sources. In 1953 Steffen\textsuperscript{34} showed that liquid In metal sources exhibited the unperturbed integral angular correlation of $^{111}\text{Cd}$, which means that this particular source was perturbation-free for at least 0.5 $\mu$sec. In a similar experiment Frauenfelder et al.\textsuperscript{35} found the same result for the integral correlation of $^{204}\text{Pb}$ involving a three times longer time range. These authors further showed that the same angular correlation was attenuated to the hard-core value in the face centered cubic environment of a $\text{Pb}_{60}$-$\text{Tl}_{40}$ alloy, while it was unperturbed in the cubic body centered phase of Pb between 235°C and 303°C. In spite of these results, almost all $g$-factor measurements with perturbed angular correlations have been carried out with ionic liquid sources.\textsuperscript{7}

It has been suggested\textsuperscript{36} that the advantage of nearly perturbation-free cubic metal sources should be exploited for all $g$-factor measurements by embedding
every isotope to be measured into metallic, cubic host lattices. This was
done, for example, in the case of $^{99}$Ru$^{36}$ and $^{100}$Rh$^{28a}$. The 235 nsec half life
of the 74.8 keV level in $^{100}$Rh allows one to observe the time spectrum for
about 1.5 μsec. For $^{100}$Rh dissolved in various cubic "non-magnetic" metals, no
attenuation of the angular correlation pattern during this time range was de-
tectable.

The reducing action of the conduction electrons brings the daughter
atom into chemical equilibrium in a very short time ($< 10^{-13}$ sec) following
the decay of the parent. Assuming there is no localized moment and neglecting
quadrupole interactions and dipolar interactions with neighboring nuclei, the
remaining perturbation arises from the contact interaction of the nuclei with
the spin magnetic moments of core s-electrons or s-like conduction electrons.$^{19}$
This is usually described in terms of the various Knight shift contributions.$^{37}$
The expected relaxation times, $T_1$, in metals are of the order of $T_1 \cdot T \approx 10^{-2}$ sec$^K$ for heavy elements, increasing to $T_1 \cdot T \approx 10^{-1}$ sec$^K$ for medium heavy and lighter
elements. Neglecting dipolar and quadrupole broadening, $T_1$ determines the
upper limit for which PAC measurements are possible in cubic metals. The time-
dependent angular correlation pattern offers an interesting alternative method
for determining $T_1$.

Thus, at room temperature, cubic metallic host lattices provide a
perturbation-free environment for lifetimes up to about $10^{-5}$ sec. This seems
to apply not only to radioactive sources, but also to thick metallic targets
in nuclear reactions,$^{28b}$ and to recoil implantation following Coulomb excitation.$^{38}$
The correction due to the paramagnetism of the conduction electrons does, in most
cases, not enter within the accuracy of PAC measurements.
3. Ferromagnetic Metallic Sources. For the measurement of magnetic moments of nuclear levels in the subnanosecond range large magnetic fields are required to obtain a detectable rotation. It was first shown by Appel and Mayer\textsuperscript{39} for Fe\textsuperscript{56} in Fe and by Deutsch, Buyrn and Grodzins\textsuperscript{40} for Sn\textsuperscript{118} and Sn\textsuperscript{120} in Fe, that internal fields can be used successfully. After-effects are not to be expected because of the metallic state. Iron is body-centered cubic and should not show any quadrupole interaction. For the face-centered cubic nickel one cannot exclude the possibility of a quadrupole perturbation,\textsuperscript{35} but it is in all probability small compared to the large magnetic dipole interaction. Thus, if the internal field is known from either NMR or Mössbauer experiments, they can be used to measure magnetic moments by PAC techniques. However, a knowledge of the field is not necessary if a sufficiently large external field is superimposed onto the internal field according to

\begin{equation}
H_{\text{eff}}^0 = H_0 + H_i
\end{equation}

If the ferromagnetic sample is saturated, there should be a linear relationship between \( g \cdot H_{\text{eff}} \) and the external field, \( H_0 \). An experimental result of this type is shown in Fig. 21. It is possible to obtain from such a measurement all quantities of interest:\textsuperscript{36} the g-factor (slope), the internal field (ordinate intercept), and the sign of the internal field (sign of slope). The internal field consists of three different contributions

\begin{equation}
H_i = H_{\text{hf}} + H_{\text{Lorentz}} + H_{\text{demag}}.
\end{equation}
The convention in the literature is to report an "internal magnetic field" which includes the Lorentz term, but which is corrected for the demagnetization field.\textsuperscript{17}

Magnetic hyperfine fields are of great value for various types of PAC work. For longer-lived nuclear states, they provide the necessary rf enhancement to make NMR experiments possible. Further, they allow the determination of magnetic moments for very short-lived levels ($\sim 10^{-12}$ sec), populated either in radioactive decay or in Coulomb excitation. With a known moment, on the other hand, the hyperfine field can be derived, but for short-lived levels no detailed information about the effective field can be expected. To obtain hyperfine fields with good accuracy time-differential or NMR measurements have to be carried out with long-lived levels. Accurate results for hyperfine fields might reveal details about the questions of magnetic field distributions, and also give some quantitative understanding of recoil or ion implantation processes.

The systematic knowledge of impurity hyperfine fields has improved considerably, mainly due to recent NMR data.\textsuperscript{20a,41} Still, there are some elements in the periodic table, for which the hyperfine field is not known. Perturbed angular distribution measurements with nuclear reactions promise to give easy access to these elements which are difficult or impossible to get into iron or nickel by chemical means. Another very promising technique is the use of isotope separators to shoot the radioactive isotope into the host lattice.
V. SUMMARY

An attempt was made to give a survey of the recent development in perturbed angular correlations. Within the scope of this presentation it was neither possible to treat all problems in detail nor to touch on all related experiments. The beautiful measurement of the hyperfine splitting of positronium by M. Deutsch and S. C. Brown was not discussed despite the fact that it was, to the knowledge of the author, the first resonance experiment with gamma-ray detection. Another experiment not mentioned here was the measurement of NMR in Ne by means of the $\beta$-asymmetry from Ne polarized in an atomic beam apparatus. While these experiments certainly fall under the subject of "radioactive detection of NMR" they are less related to perturbed angular correlations and distributions. This field of PAC has rapidly expanded and left the boundaries of conventional techniques. It now contains perturbed angular distributions obtained with Coulomb excitation and nuclear reactions, and the resonance destruction of angular distributions from statically or dynamically oriented systems. NMR experiments with radiative detection are a new stimulus which guarantees a close interplay, in the future, between PAC and resonance techniques. While, in the beginning, it was a matter of luck to find a suitable case which fulfilled all the requirements for PAC experiments, it seems that this field is slowly but systematically turning into an excellent method to investigate how nuclei are affected by their environment.

Many experiments remain to be done even before the various methods are understood in detail, for example: (1) The question of the form of hyperfine fields for sources prepared by simple melting, diffusion, recoil implantation, and ion implantation with isotope separators. (2) The determination of the sign
of the interaction frequency in NMR experiments. (3) The verification that NMR experiments in saturated samples show the domain resonance; and that the g-factor can be measured by observing a linear shift of the resonance as a function of the external field. (4) The measurement of the resonance spectrum of an electric quadrupole interaction, for example, in garnets. (5) The use of the Mössbauer effect to detect the nuclear magnetic resonance. Such a technique would have the interesting aspect that it might provide a means of detecting the Mössbauer effect in long-lived states (> 10^{-6} sec) where the present day velocity spectrometers fail. Instead of measuring a velocity spectrum, the Mössbauer effect would be recorded as a frequency spectrum. (6) The subject of "after effects" needs more investigations to obtain more quantitative results, e.g., by careful decoupling experiments, or the investigation of hyperfine fields in ferromagnetic insulators. (7) Finally, it should not be forgotten, that despite the exciting possibilities, no perturbed angular distribution measurement has yet been performed involving an isomeric level with a lifetime between 1 μsec and 1 sec.
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FIGURE CAPTIONS

Fig. 1. Comparison of the applicability of different methods to different nuclear lifetime ranges.

Fig. 2. Integral angular correlation for $^{193}$Ir in iron, measured by Auerbach, et al. The rotation of the angular correlation pattern is observed with a source polarized perpendicular to the detector plane. The rotation corresponds to $\omega r = -0.203\pm0.003$ rad.

Fig. 3. Integral angular correlation for $^{100}$Ru in iron measured by Auerbach, et al. The source was polarized with an external magnetic field of $\pm 10$ Kgauss, applied perpendicular to the detector plane. The rotation corresponds to $\omega r = -(1.78\pm0.12) \times 10^{-2}$ rad.

Fig. 4. Integral angular correlation for $^{106}$Pd in iron measured by Auerbach, et al. The source was polarized with an external magnetic field of $\pm 10$ K.G, applied perpendicular to the detector plane. The rotation corresponds to $\omega r = -(1.76\pm0.003) \times 10^{-2}$ rad.

Fig. 5. Time-differential $g$-factor measurement with a source of $^{100}$Rh in copper in an external field of $\pm 2.22$ Kgauss. In the upper part of the figure the raw data are shown and in the lower part, the corresponding ratios $R_1$ (see Eq. (10)). The full line represents the weighted least-squares fit to the data, yielding $g = +2.160\pm0.006$.

Fig. 6. Time-differential $g$-factor measurement with a source of $^{100}$Rh in silver in an external field of $\pm 5.00$ Kgauss.

Fig. 7. Time-differential angular correlation for $^{111}$Cd in nickel above the Curie point in an external magnetic field of 19.6 Kgauss. The change of the frequency for different temperatures represents a change of the negative
hyperfine field superimposed on the external field. The paramagnetic correction factors for the three temperatures are \( \beta(645^\circ K) = 0.398 \pm 0.007, \beta(651^\circ K) = 0.511 \pm 0.009, \beta(718^\circ K) = 0.879 \pm 0.011. \)

Fig. 8. Time-differential angular correlation for \(^{99}\)Ru in Ni with a polarizing field of 5.00 Kgauss. The indicated hyperfine field has been calculated assuming \( g = -0.189 \pm 0.004. \) The amplitude damping is probably caused by either a magnetic field distribution or the existence of at least two different internal fields.

Fig. 9. Time-differential angular correlation for \(^{111}\)Cd in Ni below the Curie point. The time spectra were recorded at an angle of 180° and no external polarizing field was used. The modulation pattern is characteristic of a randomly oriented magnetic interaction. The solid line represents the weighted least-squares fit to the data. The interaction frequencies for the three curves are \( \omega(4.2^\circ K) = 104.37 \pm 0.21 \text{ MHz}, \omega(454^\circ K) = 82.55 \pm 0.21 \text{ MHz}, \omega(582^\circ K) = 49.77 \pm 0.20 \text{ MHz}. \)

Fig. 10. Digital analysis of perturbed angular correlations. Displayed is the frequency spectrum of the sine transform, \( A(\nu) \), and cosine transform, \( B(\nu) \), and of the absolute function \( F(\nu) = [A(\nu)^2 + B(\nu)^2]^{1/2} \). The measurement was done in an external field of 7636±7 gauss, which gives a \( g \)-factor of \( g = 2.162 \pm 0.005. \)

Fig. 11. Cosine transform of the time spectrum obtained with \(^{100}\)Rh in three different host lattices. The shift of the resonance is due to the different Knight shift for these alloys. The external field was 8047±15 gauss.

Fig. 12. Principle of the resonance destruction of an angular correlation. The three conditions which have to be fulfilled are indicated.
Fig. 13. Sketch of the geometry of a resonance experiment with angular correlations.

Fig. 14. Resonance experiment for $^{100}$Rh in Ni at room temperature. $^{3}$ Observed was the coincidence counting rate of the 84–74.8 keV cascade at 180° as a function of frequency. $H_0 = 100$ gauss and $2H_1 \cong 3$ gauss. The internal field corresponds to a value of $197\pm2$ Kgauss.

Fig. 15. Resonance experiment for $^{100}$Rh in Fe at two different temperatures. $H_0 = 100$ gauss and $2H_1 \cong 1.5$ gauss. The corresponding internal fields are $H_1(300^\circ K) = 537.0\pm0.6$ Kgauss and $H_1(77^\circ K) = 556.5\pm0.6$ Kgauss.

Fig. 16. Experimental arrangement for a resonance experiment in statically polarized nuclei. The magnetization salt is usually a chrome-alum-glycerine slurry.

Fig. 17. Typical warm-up curves for $^{60}$Co in Fe at $\theta = 0^\circ.^{23}$ In the upper part the frequency was varied with time, and the resonance effect appears at 165.8 MHz. The lower curve was taken with a fixed frequency off resonance but with the same rf amplitude as in the upper case.

Fig. 18. Resonance effect in polarized $^{60}$Co nuclei, observed at 0° and 90° for two different alloys.$^{23}$ Both γ-ray transitions of 1.173 MeV and 1.332 MeV were taken together in the energy window. $H_0 = 1.0\pm0.5$ Kgauss. The internal field obtained from these data is $H_1 = 290.6\pm0.9$ Kgauss.

Fig. 19. Sketch of the experimental arrangement used by Sugimoto et al.$^4$ for measuring the NMR in $^{17}$F.
Fig. 20. Experimental result for the NMR in $^{17}\text{F}$ obtained by Sugimoto et al.\textsuperscript{4} The relative asymmetries $R(H)$ are plotted versus the magnetic field strength in units of the proton-resonance frequency. The solid curves represent the best fit of the data, assuming Lorentzian shape. The mean value of the magnetic moment for $^{17}\text{F}$ (not corrected for diamagnetism) obtained from these data is $\mu = 4.7200\pm0.0011$ nm.

Fig. 21. Measurement of the g-factor and the internal magnetic field of $^{99}\text{Ru}$ in Ni at room temperature.\textsuperscript{36} The sign of the slope means that the external magnetic field is opposite to the internal field. The result of the least-squares fit is $|g| = 0.184\pm10$ and $H_i = -180\pm10$ Kgauss.
Coulomb excitation

Angular correlations

Nuclear reactions

Mössbauer effect

Radioactive isotopes

Time-integral

Time-differential

Nuclear orientation

Atomic beam

10^{-12} 10^{-10} 10^{-8} 10^{-6} 10^{-4} 10^{-2} 1 10^2 sec

Sugimoto experiment

Resonance with enhancement

Resonance without enhancement

Fig. 1
Fig. 2
Fig. 4
Fig. S 100 in Cu

Coincidence rate

200 nsec

Time (arbitrary units)

Fig. 5
Fig. 6

Rh$^{100}$ in Ag  \( G = 2.1816 \pm 0.0035 \)
Fig. 7

Cd	extsuperscript{III} in Ni
T > T\textsubscript{c}
Ru$^{99}$ in Ni

4.2 °K

$H_{hf} = -216.0 \pm 1.5 \text{ kG}$

77 °K

$H_{hf} = -213.9 \pm 1.3 \text{ kG}$

Fig. 8
Fig. 9

Cd$^{111}$ in Ni,
$T < T_c$

Coincidence rate

- $4.2^\circ\text{K}$
- $454^\circ\text{K}$
- $582^\circ\text{K}$

Time (arbitrary units)

50 nsec
Fig. 10

-100$^{100}$Rh in Rh

180°

A(ν)

B(ν)

F(ν)

Frequency (MHz)

24 25 26 27

25.17
Fig. 11
Conditions:

1. $\omega_0 \tau_N \gg 1$
2. $\omega_1 \tau_N \gtrsim 1$
3. $\tau_N < \tau_{\text{relax}}$

Fig. 12
74.8 keV level
$T_{1/2} = 235 \text{ nsec}$

$^{100}\text{Rh}$ in Ni

$20^\circ \text{C}$

Fig. 14
Fig. 15

Rh$^{100}$ in Fe
77°K

300°K

Frequency (MHz)

Intensity

860 870 880 890 900

890 900 910 920 930

1.00

0.99

0.98

0.97

MUB 11913
Fig. 17
Fig. 19: Deuteron Beam (2.3 MeV)
Fig. 20
Fig. 21: 

Graph showing the relationship between $gH_{eff}$ (kG) and external field (kG) for Ru$^{99}$ in Ni.