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# Radiative Detection of Single-pulse and Spin-echo Nuclear Magnetic Resonance

*G. V. H. Wilson, D. H. Chaplin, P. Cooke, H. R. Foster and P. Lynam*

Department of Physics, Faculty of Military Studies,
University of New South Wales, Royal Military College, Duntroon, A.C.T. 2600.

*Abstract*

Experimental results on the detection of single-pulse n.m.r. of oriented radioactive nuclei are described, and a method of radiative detection of spin echoes is proposed.

## 1. Introduction

The radiative detection of n.m.r. in thermally oriented radioactive nuclei by observing the effects of a resonant RF field on the anisotropic angular distribution of the $\gamma$-ray radiations was first suggested by Bloembergen and Temmer (1953). The main advantage of radiative detection over conventional n.m.r. techniques, which detect the EMF induced in an RF coil by the precessing nuclei, would be that far fewer nuclei are required. Furthermore, such experiments should lead to new information on the fundamentals of the influence of resonant fields on the emission of radiations by nuclei (Shirley 1968). Thus, following the suggestion of Bloembergen and Temmer, several unpublished attempts were made to detect n.m.r. in nuclei that were oriented in paramagnetic dielectric materials at low temperatures. These attempts failed, probably because of the combination of nonresonant RF heating and the low thermal conductivity of the host materials. More recently, it has been realized that ferromagnetic metals offer significant advantages as host materials, e.g. large hyperfine magnetic fields generally act on the nuclei, the thermal conductivity is relatively high and, relative to the applied RF fields, there is a large ferromagnetic enhancement (Portis and Gossard 1960) of the RF fields which act on the nuclei.

The first observation of n.m.r. by radiative detection was by Matthias and Holliday (1966) using $^{60}$Co nuclei oriented in iron at a low temperature ($\sim 0 \cdot 03$ K). At this temperature the interaction energy $\mu H_e$ of the nuclei with the hyperfine magnetic field $H_e$ is comparable with $kT$, and there is then an anisotropy of $\sim 10\%$ in the emission of the $\gamma$-ray radiations. At the resonant frequency of $165 \cdot 5$ MHz, Matthias and Holliday observed only a small fractional destruction of $\sim 1\%$ of the anisotropy. The reason for this small effect was shown by Templeton and Shirley (1967) to be due to the spread ($\sim 0 \cdot 1$ T) in the hyperfine fields associated with inhomogeneous line broadening; for a single frequency, only a very small fraction of the nuclei is affected by the RF field. However, Templeton and Shirley were able to achieve large ($\sim 60\%$) resonant reductions in the anisotropy by employing a frequency-modulated RF field with a modulation amplitude that was large enough to cover the inhomogeneous broadening and a modulation frequency that was high in comparison with the spin–lattice relaxation rate. Since then, the resonances of a wide variety of radioactive nuclei in ferromagnetic host metals have been reported, and much valuable data have been obtained on their hyperfine interactions, spin–lattice relaxation processes and, in some cases, on such nuclear properties as magnetic moments and decay scheme parameters. A review of this work has been given by Stone (1976).

Despite the success of CW frequency-modulated experiments in enabling accurate measurements to be made of the interactions between the nuclei, considerable effort has been needed to reconcile theoretical and experimental results concerning the signals, e.g. the fractional resonant reductions in the radiation anisotropies. A very simple view of such experiments is that they involve a rise in the nuclear spin temperature associated with the usual (e.g. Abragam 1961) RF saturation of the nuclear polarization. However, it was pointed out (Shirley 1968) that the problem is more fundamental than this and requires consideration of the coherence in the motion of the spins in the rotating frame. Theoretical studies (Wilson *et al.* 1972; Wilson and Bosse 1974) of the motion of the spins caused by a modulated CW RF field yielded results for the signals which did not agree well with experiment. An important step was the introduction by the Monash group of single passages (Barclay *et al.* 1970, 1971, 1972). In their experiments the RF signal was swept only once through the resonance, thus making comparison between theory and experiment much simpler. Although the theoretical studies indicated that the axis of nuclear polarization should have been rotated by a single passage through the resonance, the observed signals were much less than those expected. The Oxford group (Callaghan *et al.* 1974) showed that these discrepancies could be explained by the presence of small electric quadrupole interactions. Even in a cubic host metal such interactions can arise from imperfections, from magnetostriction or from the lower (axial) symmetry introduced into the electron wavefunctions via the spin–orbit interaction and the ferromagnetic order.

Recently the Duntroon group (Foster *et al.* 1976) reported the first observation by radiative detection of single-pulse n.m.r. In these experiments a single RF frequency was used together with very large RF fields to overcome the inhomogeneous broadening effects. Probably the most detailed studies of nuclear interactions in conventional work on stable nuclei have been made using multiple-pulse spin-echo techniques. The aim of the present paper is to review the current status of single-pulse experiments and to discuss possible techniques for observing spin echoes of radioactive nuclei by radiative detection.

## 2. Theory of Single-pulse Experiments

The normalized distribution of $\gamma$-ray radiation from axially oriented nuclei may be written (Blin-Stoyle and Grace 1957) as

$$W(\theta) = 1 + \sum_{\text{even } \nu} U_\nu F_\nu B_\nu(\beta) \, \mathrm{P}_\nu(\cos\theta), \tag{1}$$

where the $\mathrm{P}_\nu(\cos\theta)$ are Legendre polynomials. The functions $B_\nu(\beta)$ describe the orientation of the parent radioactive nuclei where, for the polarization of nuclei of spin $I$ in a magnetic field $H$ at temperature $T$, we have $\beta = \mu H/kTI$. The coefficients $F_\nu$ are angular correlation functions for the observed $\gamma$-ray transition and contain coupling coefficients for the initial- and final-state spins and the angular momentum

$L$ of the observed radiation. The $U_\nu$ are angular momentum coupling coefficients expressing the attenuation of the orientation during the transitions preceding the observed $\gamma$-ray transition.

The system that has been studied most extensively by radiative detection of n.m.r. is that of $^{60}Co$ nuclei in iron. For $^{60}Co$ nuclei, we have $U_2 F_2 = -0{\cdot}4206$ and $U_4 F_4 = -0{\cdot}2429$, while $U_\nu F_\nu = 0$ for $\nu > 4$. At cobalt nuclei in iron there is a hyperfine magnetic field of $-28{\cdot}8$ T, the minus sign indicating that this field is antiparallel to the magnetization. In nuclear orientation experiments, an applied magnetic field $\sim 0{\cdot}1$–$0{\cdot}5$ T is used to align the magnetic domains of the sample to produce a common axis of nuclear polarization.

From low temperatures up to a temperature sufficiently high for the radiation to be effectively isotropic, the fractional decrease and increase respectively in the $\gamma$-ray radiation intensitites at 0° (axial) and 90° (equatorial) to the polarization axis are given (from equation 1) by

$$E(0°) = 1 - W(0°) = U_2 F_2 B_2(\beta) - U_4 F_4 B_4(\beta)\,, \tag{2a}$$

$$E(90°) = W(90°) - 1 = -\tfrac{1}{2} U_2 F_2 B_2(\beta) + \tfrac{3}{8} U_4 F_4 B_4(\beta)\,. \tag{2b}$$

For $^{60}Co$ nuclei in iron at a temperature of $0{\cdot}01$ K there is, relative to higher temperatures, a decrease of $\sim 50\%$ in the axial intensity and an increase of $\sim 20\%$ in the equatorial intensity. In nuclear orientation-n.m.r. experiments, the effect of a resonant RF field on the axial ($\theta = 0°$) intensity is usually monitored. We define the signal $S$ as the fractional resonant destruction of the axial radiation anisotropy. Hence if, as is theoretically possible in experiments with CW frequency-modulated RF fields, the populations of the nuclear Zeeman levels can be equalized then a signal of unity would be observed. On the other hand, under suitable conditions, it should be possible to interpret the effect of some experiments (such as pulsed n.m.r. or single resonant passages) in terms of a simple rotation of the polarization axis; for the usual case in which the $\nu = 2$ term dominates the anisotropy, the maximum possible signal of $1{\cdot}5$ would then be observed for a rotation through 90°.

For pulsed experiments it may be assumed that the pulse duration is short in comparison with the nuclear spin–lattice relaxation time. The analysis may then be conveniently divided into two parts: during the pulse the effects of relaxation may be neglected, while after the pulse the nuclear orientation will relax back towards lattice equilibrium. The theory of the time dependence of the orientation parameters during relaxation is generally well understood (see e.g. Gabriel 1969) and we concentrate here on the effects of the resonant RF pulses. In nuclear orientation experiments on ferromagnetic metals, the samples are magnetically saturated by an applied magnetic field $\boldsymbol{H}_A$ to produce a common axis for the orientation of the nuclei in the hyperfine magnetic fields $H_{int}$. This axis is taken to be the $z$ axis. A perpendicular linearly polarized RF field of amplitude $H_x$ is applied, and this produces a circularly polarized RF field of amplitude $H_1 = \frac{1}{2}\eta H_x$ at the nuclei. Here $\eta$ is the ferromagnetic RF enhancement factor given approximately by $\eta = H_{int}/H_A$. The resultant static field on the nuclei along the $z$ axis is $H_0 = H_{int} \pm H_A$, the sign being that of the hyperfine field. It has been shown previously (Barclay *et al.* 1971) that, where the only external interactions of the nuclei are with magnetic fields, the orientation parameters $B_\nu$ are invariant with respect to a quantization axis which rotates so as to remain parallel to the instantaneous nuclear magnetization. Furthermore, quantum mechanical calculations of the motion of the components of the nuclear magnetization then lead to results that are identical to those derived from simple solutions of the classical torque equation (Rabi *et al.* 1954).

Because of the need to overcome the effects of inhomogeneous line broadening, it is necessary to use large RF fields in the pulsed experiments, and we assume here that the small quadrupole interactions of the nuclei in cubic ferromagnetic metals (Callaghan *et al.* 1974) may be neglected. As above, the effects of the pulse upon the radiation anisotropy are then determined by calculating the motions of the magnetizations of all of the radioactive nuclei in the magnetic fields. This motion may be best considered by the usual transformation (see e.g. Abragam 1961) to a frame of reference which rotates about the $z$ axis at the RF frequency $\omega$. In this frame an effective field $\boldsymbol{H}_{\text{eff}}$ acts on the nuclei and is given by

$$\boldsymbol{H}_{\text{eff}} = (H_0 - \omega/\gamma)\hat{\boldsymbol{z}} + H_1\hat{\boldsymbol{x}}. \tag{3}$$

The motion of the nuclear magnetization $\boldsymbol{M}$ in the laboratory frame will then be precession about $\boldsymbol{H}_{\text{eff}}$ at a frequency $\omega_{\text{eff}} = \gamma H_{\text{eff}}$ together with rotation at frequency $\omega$ about the $z$ axis. The latter rotation is too fast to be resolved by the $\gamma$-ray detectors and, by averaging over the radiofrequency period (i.e. over all phase angles $\phi$) and employing the spherical harmonic addition theorem, it follows that the angular distribution of the $\gamma$-ray radiation at time $t$ will be given by

$$W(\theta, t) = 1 + \sum_{\text{even } \nu} U_\nu F_\nu B_\nu(\beta)\, \mathrm{P}_\nu(\cos\theta)\, \mathrm{P}_\nu(\cos\alpha), \tag{4}$$

where $\alpha$ is the instantaneous angle between $\boldsymbol{M}$ and the $z$ axis (in either frame). The simplest case, and that which is normally assumed in treatments of conventional pulsed n.m.r., is that the RF is switched on sufficiently rapidly at $t = 0$ so that $\boldsymbol{M}$ precesses about $\boldsymbol{H}_{\text{eff}}$ in a cone which passes through the $z$ axis. Hence the cone angle $\theta_c$ is given by

$$\tan\theta_c = \omega_1/(\omega_0 - \omega), \tag{5}$$

where $\omega_1 = \gamma H_1$. At resonance, in the rotating frame, $\boldsymbol{M}$ precesses in the $yz$ plane about the field $H_1\hat{\boldsymbol{x}}$ with frequency $\omega_1$. The 90° and 180° pulses of conventional n.m.r. are then obtained for pulse widths of $\pi/2\omega_1$ and $\pi/\omega_1$ respectively. The signal immediately after a pulse of width $\tau$ will be given by equation (4) with $\alpha = \omega_1\tau$, leading to

$$S_2 = 1 - \mathrm{P}_2(\cos\omega_1\tau) \tag{6}$$

for the case of a dominant $\mathrm{P}_2$ term in the $\gamma$-ray anisotropy. Hence signals varying between 0 and 1·5 as a function of $\tau$ are expected. Apart from the possibility of small quadrupole interactions, two effects will tend to reduce these signals and to smear out the variation in equation (6). These are the inhomogeneous broadening of the resonance, and the distribution of RF field amplitudes associated with the geometry of the RF coil system and with the effects of RF eddy currents in the sample.

Assuming that there is an inhomogeneous broadening with a probability distribution of resonant frequencies $P(\omega_0)$, leads us to a signal

$$S_2 = 1 - \int_0^\infty \mathrm{P}_\nu(\cos\alpha)\, P(\omega_0)\, \mathrm{d}\omega_0, \tag{7}$$

where (Abragam 1961) the angle $\alpha$ is given by

$$\cos\alpha = 1 - 2\sin^2(\theta_c)\sin^2(\tfrac{1}{2}\omega_{\text{eff}}\,t). \tag{8}$$

If we then assume a gaussian probability distribution

$$P(\omega_0 - \bar{\omega}_0) = \frac{1}{\Delta}\left(\frac{\ln 2}{\pi}\right)^{\frac{1}{2}} \exp\left(\frac{-(\ln 2)(\omega_0 - \bar{\omega}_0)^2}{\Delta^2}\right), \tag{9}$$

where $\Delta$ is the halfwidth at half-maximum height, we obtain

$$S_2 = 1 - \int_{-\infty}^{\infty} P(\omega_0 - \bar{\omega}_0)\,\mathrm{P}_2(\cos\alpha)\,\mathrm{d}(\omega_0 - \bar{\omega}_0). \tag{10}$$

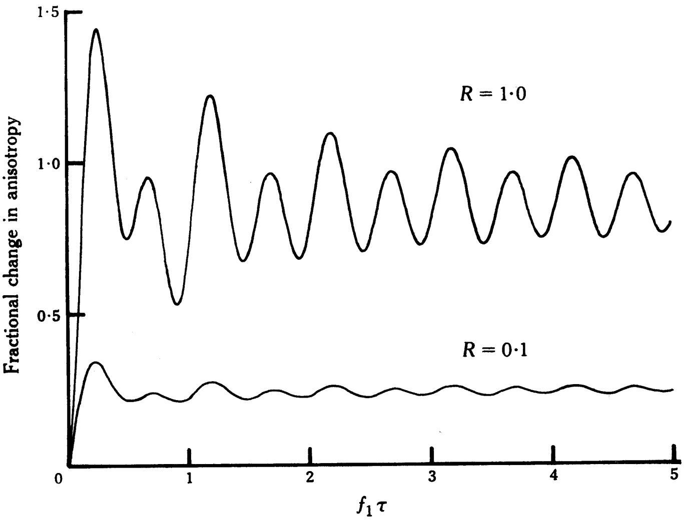


**Fig. 1.** Theoretical dependence of the signal on $f_1\tau$ (where $f_1 = \omega_1/2\pi$) for an inhomogeneously broadened line. The ordinate is the fractional change in anisotropy of the $\gamma$-ray radiation. The two curves are for the indicated values of $R = \omega_1/\Delta$.

The variation of the signal with $f_1\,\tau$ (where $f_1 = \omega_1/2\pi$) for two values of $R = \omega_1/\Delta$ is shown in Fig. 1. For $R \gg 1$ (not shown) the signal oscillates between 0 and 1·5 at a frequency $2f_1$, as in equation (6). For $R = 0{\cdot}1$, Fig. 1 shows the reduced signal amplitude and the damping of the oscillation which occur when $\omega_1$ is not large compared with $\Delta$. However, it should be noted that the frequency of the oscillations still remains close to $2f_1$.

One important difference between conventional and radiative detection of single pulse experiments should be noted. In conventional experiments, after a 90° pulse, the RF EMF induced in a pickup coil by the precessing magnetization is observed immediately, and this EMF decays in a time $\sim 1/\Delta$ as the inhomogeneous broadening leads to phase incoherence between the precessions of different nuclei. This is the free induction decay which is generally fast in comparison with the spin–lattice relaxation time $T_1$. In radiative detection, because the $\gamma$-ray detection system does not time resolve the effects of precession about the $z$ axis, no change in signal will be produced by the decay from phase coherence to incoherence. Instead the γ-ray anisotropy will be observed to relax from the value it had immediately after the pulse to its initial value as the nuclei return to lattice equilibrium via spin–lattice relaxation.

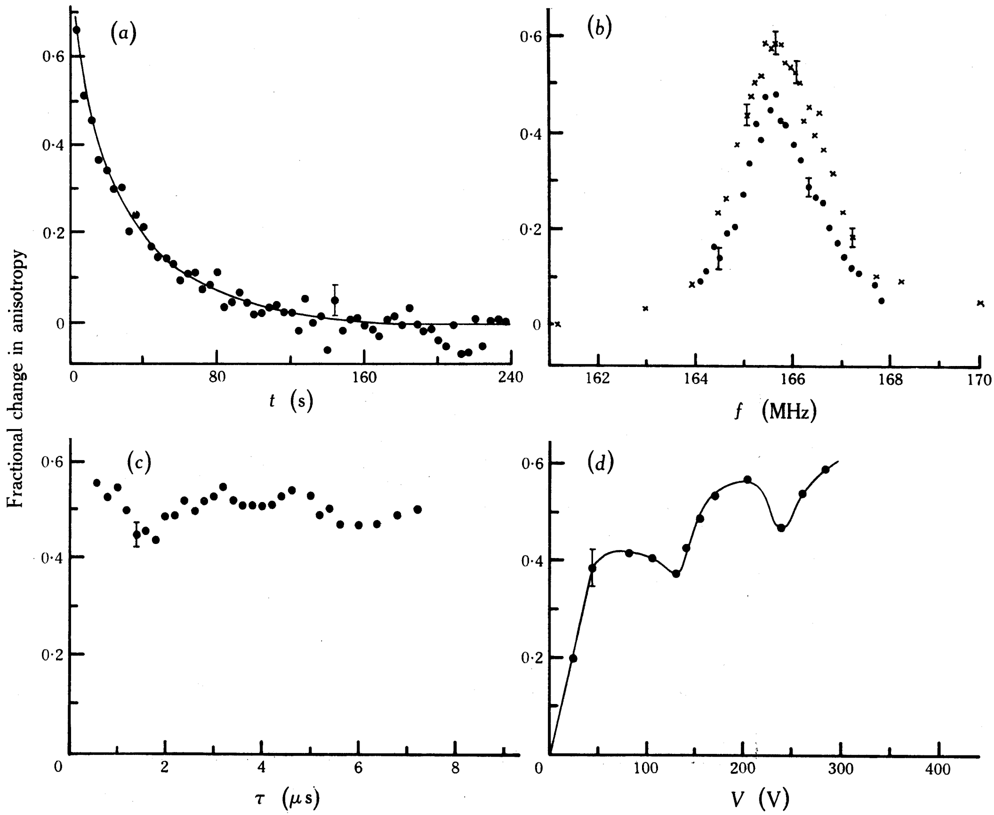


**Fig. 2.** Dependence of the fractional change in the γ-ray radiation anisotropy for $^{60}$Co nuclei in iron at 0·01 K on:

(a) time $t$ for an RF voltage amplitude $V = 300$ V and pulse width $\tau = 2\ \mu$s;
(b) RF frequency $f$ for $\tau = 10\ \mu$s and $V = 175$ (circles); and 275 V (crosses);
(c) pulse width $\tau$ for $V = 130$ V;
(d) RF voltage amplitude $V$ for $\tau = 2\ \mu$s.

## 3. Experimental Details and Results

Our previous report (Foster *et al.* 1976) of radiative detection of pulsed n.m.r. was for a sample which had been prepared by melting some iron with carrier-free $^{60}$Co activity, a small piece of which had been cold rolled to a thickness of 2 μm. Signals as high as $S_2 \approx 0 \cdot 37$ were recorded with single-pulse experiments. However, the detailed interpretation of these experiments was made difficult by the presence of a large electric quadrupole interaction, which was indicated by a strong dependence of the signals from single-passage experiments upon the direction of frequency sweep. The single-pulse experiments reported here were performed on a sample prepared by diffusing a few μCi ($\sim 10^5$ Bq) of $^{60}$Co activity into the surface of a 200 μm thick disc of iron. The diffusion temperature and time were 1103 K and 4 min respectively, leading to an expected mean diffusion depth $\sim 0 \cdot 7$ μm. Single-passage experiments on this sample indicated that any quadrupole interactions were very weak, with nuclear interaction frequencies $\lesssim 10$ kHz. The much larger quadrupole interactions of the first sample were presumably associated with strains produced by the cold rolling.

In the single-pulse experiments a gated RF signal generator was used together with a 250 MHz bandwidth amplifier capable of producing 1 kW RF pulses with $0\cdot3$ $\mu$s risetimes. Fig. 2*a* shows the time dependence of the $\gamma$-ray radiation anisotropy for a pulsed experiment with a peak of voltage $V = 300$ V and a pulse width $\tau = 2$ $\mu$s. The figure shows a maximum change $S_2 = 0\cdot65$ in the anisotropy followed by a decay back to the equilibrium anisotropy by spin–lattice relaxation. The dependence of the signal upon the RF frequency for two values of the voltage amplitude of the RF field is shown in Fig. 2*b*. For $V = 275$ V the halfwidth is $1\cdot05$ MHz. From experiments using small frequency-modulated RF fields, a halfwidth of $0\cdot5$ MHz was observed. The broadening of the spectra obtained from pulsed experiments with large RF fields is caused by power broadening associated with the large values of $\omega_1$. From the results shown in Fig. 2*b* we obtain an approximate value $\omega_1/2\pi V \approx 1\cdot6\ \mathrm{kHz\,V^{-1}}$.

Figs 2*c* and 2*d* show the dependence of the signal on $\tau$ for $V = 130$ V, and on $V$ for $\tau = 2$ $\mu$s respectively. Noting that $H_1$ (and hence $\omega_1$) are proportional to $V$, both of these figures show the expected oscillatory dependence of the signal upon $\omega_1 \tau$. From this an RF amplitude calibration was obtained for our system of $\omega_1/2\pi V = 2\cdot2 \pm 0\cdot2\ \mathrm{kHz\,V^{-1}}$. It should be noted that to obtain an accurate estimate of this important parameter of the RF field in the vicinity of the nuclei in such experiments has previously been difficult, as this involved the geometry of the coils, the sample and the neighbouring cryostat components, together with details of the ferromagnetic enhancement. The present pulsed experiments are the first radiative detection experiments to permit this calibration to be made from observations of the RF precession.

From the deduced value of $\omega_1$ for the data of Fig. 2*c* it follows that $\omega_1/\Delta = 0\cdot48$. Comparison with the calculated curves, such as those in Fig. 1, indicates that the observed oscillations are not as pronounced as expected. Our current work (unpublished) is aimed at establishing the cause of this discrepancy by reducing the distribution of the RF field amplitudes and by employing shorter pulse risetimes.

## 4. Proposed Spin-echo Experiment

The most important applications of conventional spin-echo n.m.r. experiments are in the study of relaxation effects. By employing a variety of pulse sequences both spin–lattice and spin–spin relaxation times may be measured. The simplest, and most widely used, sequence is that of a 90° pulse followed by a 180° pulse. If $t_1$ is the time interval between the two pulses then, for values of $t_1$ which are greater than $1/\Delta$ but not too large in comparison with the relaxation times, a spin echo will occur at a time $t_1$ after the second pulse. The spin echo is the result of a coherence in the precession of the nuclei about the $z$ axis and lasts for a time $\sim 1/\Delta$. We discuss here possible techniques for observing spin echoes by radiative detection.

In conventional experiments the echo is detected via the EMF induced in a pickup coil by the temporary coherence of the nuclear precession. However, as described in Section 2, nuclear orientation experiments have not yet been performed with a suitable time-sorting equatorial detection system. The rotation of the angular distribution of the $\gamma$-ray radiation would occur at twice the Larmor frequency. Although the necessary time resolution ($\lesssim 1$ ns) is now obtainable, such a system must be ruled out at present on the basis of counting statistics. The echoes will only have durations $\sim 1\ \mu$s with an interval of order $T_1$ (typically seconds for ferromagnetic metals at the required low temperatures). The difficulty is further increased because of limits on the sample activity which are necessary to avoid radioactive heating effects.

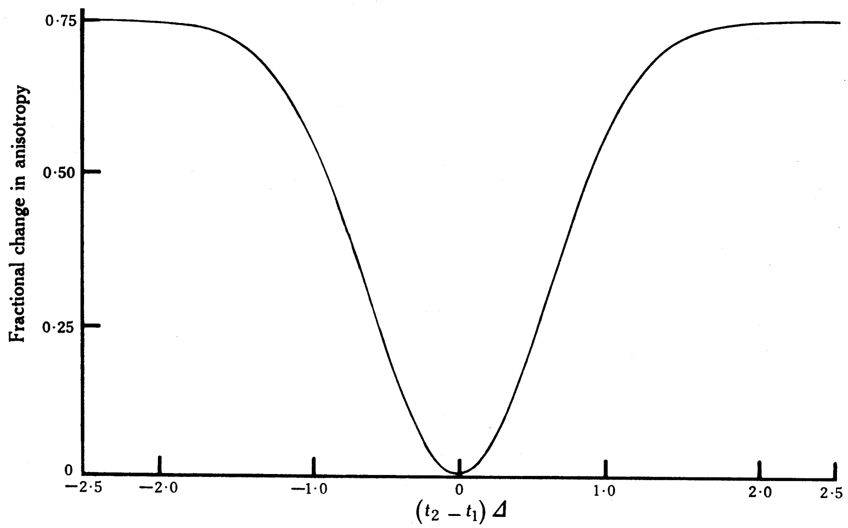


**Fig. 3.** Calculated dependence of the fractional change in $\gamma$-ray radiation anisotropy after a triple-pulse (90°, 180°, 90°) sequence upon $(t_2-t_1)\Delta$. This is the theoretical spin-echo nuclear orientation lineshape.

We now propose a technique in which an additional 90° pulse may be applied to indicate the occurrence (or otherwise) of an echo. To the simple 90° — 180° sequence let us add this final 90° pulse at a time $t_2$ after the 180° pulse. Provided that $t_1$ and $t_2$ are both $\ll T_1$, the nuclei will all be precessing in the $xy$ plane before the third pulse. At the peak of an echo the nuclei will be polarized along the $-y$ direction in the Larmor frame, while after the third pulse they will be rotated to their initial direction in the laboratory frame. Hence if we choose $t_2 = t_1$, so that the third pulse occurs at the echo time, no change in the anisotropy of the $\gamma$-ray radiation will be produced by the triple pulse sequence—the sum of all three pulses will be equivalent to a rotation of the initial nuclear orientation through 360°. If on the other hand the third pulse is applied when there is no echo, the final result will be (in the laboratory frame) a plane of randomly oriented nuclei precessing about the $z$ axis. This will lead to a signal $S_2 = 0{\cdot}75$. The spin echo should therefore be observable by measuring the change in radiation anisotropy produced by the triple pulse sequence as a function of the time $t_2$. The signal will be given by

$$S_2 = 1 - \int_{-\infty}^{\infty} P(\omega_0 - \bar{\omega}_0)\, \mathrm{P}_2(\cos\beta)\, \mathrm{d}(\omega_0 - \bar{\omega}_0)\,, \tag{11}$$

where $\beta = (t_2 - t_1)(\omega_0 - \bar{\omega}_0)$. This signal is plotted against $(t_2 - t_1)\Delta$ in Fig. 3 for the gaussian line-broadening distribution (9).

An important application of the proposed technique would be to separate the effects of spin–spin and spin–lattice relaxation in dilute radioactive alloys. In the above analysis it is assumed that each pulse produces the desired rotation of the spins of all nuclei. However, the successful application of the technique will require improvements to be made in the resolution of single-pulse rotations as discussed in Section 3.

## 5. Conclusions

The radiative detection of rotations of nuclear polarization in single-pulse n.m.r. experiments has been demonstrated. It has been shown that problems of counting statistics and time resolution will not prevent the radiative detection of spin echoes if the signals from triple pulse sequences are measured. However, improvements in the definition of rotations from single pulses will be necessary.

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