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## NUCLEAR MAGNETIC RESONANCE IN POLARIZED NUCLEI\*

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The nuclear ferromagnetic resonance of  $^{60}\text{Co}$  in Fe has been detected by observing the effect of a rf field on the angular distribution of the  $\gamma$  radiation emitted by polarized  $^{60}\text{Co}$  nuclei at temperatures of about  $0.03^\circ\text{K}$ . At resonance, rf induced transitions partially randomize the Boltzmann distribution, which is set up by the low temperature and the large hyperfine field for Co in Fe, in the  $^{60}\text{Co}$  ground state. The randomization has a destructive effect on the angular distribution of the emitted  $\gamma$  rays, which can be detected by observing the counting rate at a specific angle as a function of frequency. The enhancement of the rf amplitude in a ferromagnet<sup>1</sup> was necessary to keep the rf power at a sufficiently low level in order to avoid a rapid warm-up of the sample.

This experiment was part of a more general program of our group in which we are exploring the possibilities of performing nmr experiments in radioactive states, using the emitted radiation pattern to detect the resonance. From the detection point of view all methods can be used that involve emitted radiation with an anisotropic angular distribution (or correlation) which can be destroyed by the rf field at resonance. Three different types of experiments are possible: (1) nuclear orientation at low temperatures, (2) angular correlations, and (3) angular distributions produced by nuclear reactions. Apart from the different experimental techniques, the three methods apply to dif-

ferent lifetime regions of the nuclear state. For short lifetimes ( $\tau_N < 10^{-6}$  sec) both (2) and (3) can be used. For lifetimes between  $10^{-6}$  and 1 sec, only method (3) would apply, and for states with  $\tau_N > 1$  sec, low-temperature orientation is applicable. The given time limits are approximate and might in some cases change as much as an order of magnitude. In a remarkable experiment of type (3), Sugimoto *et al.*<sup>2</sup> proved that the polarization of nuclei produced by a nuclear reaction can be preserved up to 10 sec or longer at room temperature and subsequently used to detect the nuclear magnetic resonance. The resonance destruction of an angular correlation was also shown to be possible<sup>3</sup> by making use of the ferromagnetic enhancement of the rf amplitude. An experiment of type (1) was suggested by Bloembergen and Temmer<sup>4</sup> in 1953, but to our knowledge, no successful experiment has been carried out along this line. The reason for this was probably the large heating effect of the rf field.

In an attempt to investigate the suggestion of Bloembergen and Temmer further, we chose again to take advantage of the rf amplitude enhancement in a ferromagnetic sample. For polarized foils the rf field,  $H_1$ , is amplified according to the relation<sup>1</sup>  $2H_1^{\text{eff}} = (1 + H_{\text{hf}}/H_0)H_1$ , where  $H_0$  is the polarizing external field and  $H_{\text{hf}}$  is the hyperfine field. In this way, it was possible to keep the externally applied rf pow-

er sufficiently low so that an adequate warm-up of the sample was obtained. For the first experiment of this type we chose radioactive  $^{60}\text{Co}$  for which both the magnetic moment and the hyperfine field in iron are known accurately, thereby making the search for the resonance easier, as we anticipated a fairly small line-width.  $^{60}\text{Co}$  emits two  $\gamma$  rays of 1.173 and 1.332 MeV in cascade, both of which have negative anisotropy and were therefore taken together in the window of the single-channel analyzer to give a larger total counting rate.

The low temperature ( $0.02^\circ\text{K}$ ) was reached by adiabatic demagnetization of a chrome-alum-glycerine slurry. The sample was in thermal contact with this salt by means of copper fins which reached into the demagnetization salt.<sup>5</sup> The sample was a thin ( $\sim 10^{-4}$  cm) Fe foil about  $3 \times 3$  mm<sup>2</sup> onto which the activity was electroplated. The activity was then diffused (alloy I at  $950^\circ\text{C}$  for 80 hr, alloy II at  $950^\circ\text{C}$  for 48 hr) into the foil and soldered with Wood's metal onto the copper fins. The polarizing field,  $H_0$ , was parallel to the plane of the foil and was produced by a pair of Nb rings located on either side of the foil. The rf field,  $H_1$ , oriented parallel to the plane of the foil but perpendicular to the polarizing field  $H_0$ , was fed in by a two-turn coil in the He bath around the cryostat.

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

$$W(\theta) = \sum_{k \text{ even}} B_k U_k F_k P_k(\cos\theta)$$

with the usual definition of the coefficients  $B_k$ ,  $U_k$ , and  $F_k$ .<sup>5,6</sup> The orientation parameters  $B_k$  are functions of the Boltzmann distribution, depending on the magnetic moment,  $\mu$ , effective magnetic field,  $H_{\text{eff}}$ , and the temperature. Resonance-induced transitions consequently alter the orientation parameters  $B_k$ ; resonance is observed as a change in radiation intensity at a fixed angle  $\theta$ . This effect can be seen in the upper part of Fig. 1, where the warm-up curve for  $\theta = 0^\circ$  is shown as a function of frequency, which was varied at constant time intervals. The resonance effect is clearly visible at about 165.8 MHz. To check whether this peak is really a frequency effect, we measured the warm-up curve for the same temperature interval and the same rf amplitude but at a fixed frequency off resonance. The result was negative, as shown in the lower part of Fig. 1. To improve statistics we added together the results

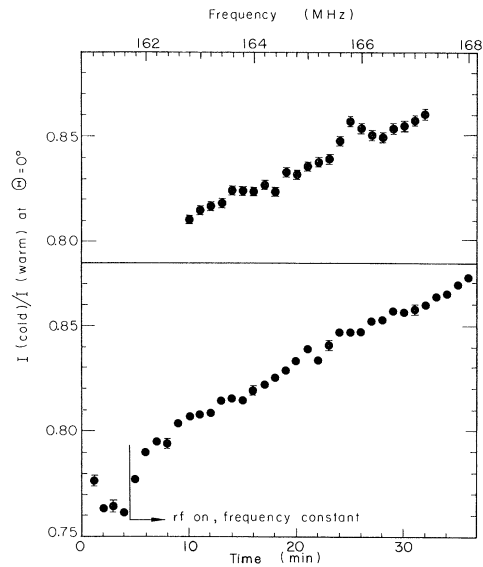


FIG. 1. Warm-up curves for  $^{60}\text{Co}$  in Fe (alloy II) at  $\theta = 0^\circ$ . In the upper part the frequency was varied with time, and the resonance effect appears at 165.8 MHz. The lower curve was taken with a fixed frequency off resonance but with the same rf amplitude as in the upper case.

of different demagnetizations, and the final result for two different alloys is shown in Fig. 2. The data in the left part of this figure (alloy I) are the sum of four demagnetizations, in which the frequency range was scanned twice per demagnetization. The data on the right (alloy II) are the result of two demagnetizations with one frequency scan after each. Although the effect at  $90^\circ$  is smaller, the opposite sign compared with  $0^\circ$  is apparent. The observed line-widths of about 0.8 MHz (alloy I) and 0.5 MHz

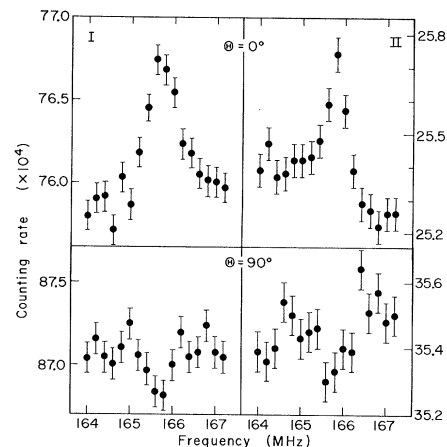


FIG. 2. Resonance effect at  $0^\circ$  and  $90^\circ$  for two different alloys (I and II) of  $^{60}\text{Co}$  in Fe.

(alloy II) are of the same order of magnitude as observed in nuclear ferromagnetic resonance. The slightly larger linewidth of the resonance in alloy I is in all probability caused by a larger inhomogeneity of the polarizing field,  $H_0$ , due to the fact that alloy I was larger. Also, the second frequency scan after a demagnetization sees a slightly displaced resonance compared with the first scan which is probably because of a change of the trapped flux in the Nb rings. To draw any conclusions from the observed linewidth, one would have to use a better-controlled external polarizing field with greater homogeneity. This would also provide the possibility of observing the change of the resonance frequency as a function of the external field and thereby measuring the  $g$  factor of the nuclear state directly.

The average value of the resonance frequency taken from both alloys is  $165.75 \pm 0.15$  MHz. With a magnetic moment of  $\mu = 3.754 \pm 0.008$  nm,<sup>7</sup> this gives an effective magnetic field of  $|H_{\text{eff}}| = 289.6 \pm 0.7$  kG. Assuming that the demagnetization factor for a thin foil is negligible we obtain, after correcting for the polarizing field ( $1.0 \pm 0.5$  kG), the value

$$|H_i| = 290.6 \pm 0.9 \text{ kG}$$

for the internal magnetic field of very dilute Co in Fe (less than 1 in  $10^3$  atoms) at  $0.03^\circ\text{K}$ . This agrees well with the value of 289.7 kG measured by nmr for 1% Co in Fe<sup>8</sup> and 4 to 17% Co in Fe.<sup>9</sup>

To conclude, it can be said that the radiative detection of nmr in polarized nuclei is proved to be possible. Compared with conventional nuclear polarization measurements,<sup>6</sup> this method offers at least an order of magnitude higher accuracy which also makes it possible to determine the nuclear  $g$  factor directly by varying the external polarizing field. Further, it is independent of any precise knowledge of the temperature scale. It should be noted that the conventional nuclear-orientation technique using  $\gamma$  rays measures an energy  $|\mu H_{\text{eff}}|$  while the present method gives a frequency  $|gH_{\text{eff}}\mu_N/$

$h|$ . Combining both techniques would determine the spin of the nuclear state.<sup>10</sup> The technique will also prove valuable for studies of the magnetic hyperfine fields of impurities in ferromagnetic lattices; it measures to a good approximation the hyperfine field at zero degree 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.

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<sup>4</sup>N. Bloembergen and G. M. Temmer, *Phys. Rev.* **89**, 883 (1953).

<sup>5</sup>For detailed information on this technique see G. A. Westenbarger and D. A. Shirley, *Phys. Rev.* **138**, A161 (1965).

<sup>6</sup>D. A. Shirley, Lawrence Radiation Laboratory Report No. UCRL-16764 (to be published).

<sup>7</sup>I. Lindgren, in Perturbed Angular Correlations, edited by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1964), p. 385.

<sup>8</sup>Y. Koi, A. Tsujimura, T. Hihara, and T. Kushida, *J. Phys. Soc. Japan* **16**, 1040 (1961).

<sup>9</sup>E. Simanek and Z. Sroubek, *Czech. J. Phys.* **B12**, 202 (1962).

<sup>10</sup>We are grateful to Dr. D. A. Shirley for calling this to our attention.