Nuclear Magnetic Resonance of Aligned Radioactive Nuclei*

N. BLOEMBERGEN, *Harvard University, Cambridge, Massachusetts*
AND
G. M. TEMMER, *National Bureau of Standards, Washington, D. C.*
(Received December 23, 1952)

IT occurred to one of us (G.M.T.) that the anisotropy of radiation from radioactive nuclei observed recently[1–4] might be used with advantage for a precise determination of the nuclear gyromagnetic ratio of radioactive isotopes. An experiment is proposed where the minimum number of nuclei necessary to detect magnetic resonance is not determined by the signal-to-noise ratio of radiofrequency equipment, but solely by the statistics of the counters monitoring the radioactive decay. Thus, only minute amounts of the radioactive isotope are needed.[5]

Suppose that in some way a certain alignment of radioactive nuclei in a crystal has been established. When this crystal is subjected to a radiofrequency field of the proper frequency, so that it will induce transitions to levels with other spatial quantization of the radioactive nuclei, the observed radioactive anisotropy will drop or disappear. This effect will occur regardless of the order of magnitude of the spin-spin relaxation time T_2 and the spin-lattice relaxation time[6] T_1. If both T_1 and T_2 are large compared to the inverse of the "nutation frequency" $1/\gamma H_{\rm rf}$, the nuclear spins will oscillate as free spins between the connected spin levels.[7] If $T_2 \ll 1/\gamma H_{\rm rf}$, but $\gamma^2 H_{\rm rf}^2 T_1 T_2 \gg 1$, the nuclear spins will lose their coherence. They will be distributed evenly over the connected spin levels. If finally $\gamma^2 H_{\rm rf}^2 T_1 T_2 \gg 1$, the power absorbed by the radioactive nuclear spins will serve to heat the other degrees of freedom in the crystal. Since a radiofrequency field of a few gauss amplitude will induce transitions at a rate of 10^4 or 10^5 per second, the absorption would be sufficient to increase the entropy of all spins in the crystal in a few seconds, even if the radioactive nuclei occur only in a relative concentration of one part in ten thousand. Spin levels with a much larger splittings, which would require more energy to increase their entropy, need not be considered, since the nuclear alignment will disappear at temperatures, where the contribution to the specific heat of these larger splittings is still negligible. There is absolutely no limitation on the concentration of the radioactive nuclei if $\gamma^2 H_{\rm rf}^2 T_1 T_2 \gg 1$.

Under all circumstances the radioactive anisotropy will be drastically reduced within a few seconds, after the application of a radiofrequency field of sufficient amplitude, at the resonance frequency of the radioactive nuclei. This frequency can thus be determined with the usual precision of the nuclear magnetic resonance in solids. The proposed method, which requires no refined high frequency techniques, is based upon a heating effect, as in Gorter's original proposal of magnetic resonance.[8] The crystal at very low temperatures constitutes a very sensitive calorimeter, the radioactive anisotropy serving as the appropriate thermometer.

If nuclear polarization is obtained for nuclei in nonmagnetic ions, the experiment can be carried out in a known external magnetic field and the nuclear magnetic moment can be measured directly, provided quadrupolar effects can be neglected. So far, nuclear alignment has been obtained only for nuclei in magnetic ions,[1–4] oriented by the strong field from the electron spins. In this case, with zero external field, the magnetic moment can be compared with that of another stable isotope, or the internal field must be calculated from theoretical considerations.[9] The magnetic moment of Co^{60} can be compared with that of Co^{59} in the same salt.[10] The resonance of the Co^{59} nuclei could, of course, also be detected by the heating of the crystal and the disappearance of the radioactive anisotropy of Co^{60}. Bleaney[3] and Gorter[8] have obtained a value of 3.2 ± 0.3 nuclear magnetons for Co^{60} from the observed magnitude of the anisotropy at a known temperature. This approximate value would facilitate the search for the precise magnetic resonance of Co^{60}. A rough calculation indicates that our proposed experiment could be carried out on the samples used by Bleaney and Gorter.

A complication may arise in magnetic salts from nonresonant electronic spin-spin absorption. During the search for the nuclear resonance, the crystal may be warmed up by this spurious absorption. It is true that nuclear alignment is achieved in crystals which are magnetically dilute, so that the nuclear resonance frequency is high compared to the spin-spin frequency. In other words, the hyperfine structure is well resolved. Nevertheless, the absorption in the tail of the spin-spin interaction might still be prohibitively large. If we assume with Broer[11] a Gaussian shape for the spin-spin relaxation, the absorption in the tail will be negligible. It would be interesting in its own right to investigate the nonresonant high frequency absorption in dilute magnetic crystals at very low temperatures. Any of the low temperature thermometers,[12] including the radioactive anisotropy could be used for this purpose.

The experiment is being initiated by the Cryogenics Section of the National Bureau of Standards.

* First reported at the meeting of the American Association for the Advancement of Science, December, 1952, in St. Louis, Missouri.

[1] Daniels, Grace, and Robinson, Nature **168**, 780 (1951).
[2] Gorter, Poppema, Steenland, and Beun, Physica **17**, 1050 (1951).
[3] Bleaney, Daniels, Grace, Halban, Kürti, and Robinson, Phys. Rev. **85**, 688 (1952).
[4] Gorter, Tolhoek, Poppema, Steenland, and Beun, Physica **18**, 135 (1952).
[5] Compare the magnetic resonance in positronium observed by M. Deutsch and S. C. Brown, Phys. Rev. **85**, 1047 (1952).
[6] In order to first establish nuclear alignment, T_1 should not be too long.
[7] See, e.g., N. Bloembergen, thesis, Leiden, 1948 (unpublished).
[8] C. J. Gorter, Physica **3**, 995 (1936).
[9] R. J. Elliott and K. W. H. Stevens, Proc. Phys. Soc. (London) **64**, 205 (1951).
[10] K. D. Bowers (unpublished).
[11] L. J. F. Broer, Physica **10**, 801 (1943).
[12] Steenland, de Klerk, and Gorter, Physica **15**, 711 (1949); D. de Klerk, thesis, Leiden, 1948 (unpublished); M. J. Steenland, thesis, Leiden, 1952 (unpublished).

Exchange Magnetic Moments of Nuclei with Almost Closed Shells of Nucleons

RICHARD W. COLE
Cornell University, Ithaca, New York
(Received December 30, 1952)

THE measured magnetic moments of H^3 and He^3 show that there is an exchange contribution to the magnetic moments of nuclei. Do the measured magnetic moments of heavier nuclei support the same conclusion? Several papers[1] seem to give an affirmative answer. Ross[2] implies that more research is needed before the question can be answered. A detailed study of a few nuclei seems to confirm Ross.

According to the individual-particle model, some nuclei have closed shells of nucleons. Nuclei which have one nucleon outside closed shells or one nucleon missing from a closed shell may be called "closed shell (±1) nuclei." If the individual particle model were exact and exchange effects contributed nothing, the magnetic moments of closed shell (±1) nuclei would lie on the Schmidt curves. Feenberg[3] has noted that the measured magnetic moments of closed shell (±1) nuclei seem to lie nearer to the Schmidt curves than those of other nuclei.

Osborne and Foldy[4] have suggested several operators for the exchange magnetic moment. Some of the operators may be eliminated because they do not have equal and opposite values in H^3 and He^3. An argument about time reversal,[5] which is not conclusive,[6] eliminates other operators. The Osborne-Foldy operators $\mathbf{M}_l$, (c), and (d) remain. The writer has omitted (d) and assumed that the exchange magnetic moment operator is

$$\mathbf{M}_x = \mathbf{M}_l + C \sum_{u,v} f(r_{uv}) \{\boldsymbol{\tau}_u \times \boldsymbol{\tau}_v\}_z \{\boldsymbol{\sigma}_u \times \boldsymbol{\sigma}_v\}$$
$$= \mathbf{M}_l + \mathbf{M}_\sigma.$$

The constant C was adjusted to fit the measured magnetic moments of H^3 and He^3.