that this may reduce the magnitude of the polarization calculated below by roughly a factor 2. The microwave field through the $\mathcal{H}_{rf}$ term induces the transitions ($\Delta m=\pm 1$, $\Delta M=0$) by the rf component perpendicular to H_z. These are the usual $2I+1$ paramagnetic resonance hfs lines. The rf component parallel to H_z induces the forbidden transitions $\Delta(m+M)=0$. These show up as $2I$ lines between the main lines. Let the rf field be polarized parallel to H_z and great enough to saturate the states b and j at resonance, i.e., make their populations equal. Then the unnormalized relative populations of the states are as shown in Fig. 1, where $2\delta=h\nu/kT$, and we have expanded the Boltzmann exponential, keeping only first order terms. The relatively small energy difference between adjacent M states is neglected. A similar situation occurs when the c and i levels are saturated, etc. Thus in general one expects $2I$ dc magnetic field values for which a fixed microwave frequency ν will induce a partial nuclear polarization. This polarization may be detected, for example, by observing the γ-ray anisotropy $\epsilon=1-\mathcal{W}(0)/\mathcal{W}(\pi/2)$ if the nuclei are radioactive. The relative magnitudes of the anisotropy lines can, in principle, uniquely determine the decay scheme. The lines form an antisymmetric pattern about their center point; this is due to the fact that the saturation of lines b and j, for example, give a population distribution which is inverted relative to that obtained by saturating e and g. As Abragam[4] points out, saturation of levels a and j will also give a nuclear polarization via the relaxation transitions b to j. The resulting anisotropy is of comparable magnitude and opposite in sign to that obtained by the direct saturation of levels b and j. The magnitude of the anisotropy depends on the decay scheme and is of order $\epsilon\sim(h\nu/kT)(2I+1)^{-1}$ for the saturation of a single line. This may be several percent at 3-cm wavelength and 1.5°K.

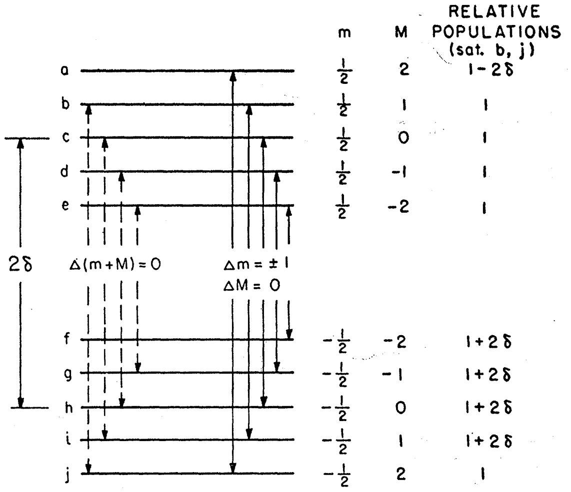

FIG. 1. Energy levels and transitions.

This method and related ones may be termed "high"-temperature dynamic polarization methods in contrast to the experiments of the Oxford and Leiden groups in which radioactive nuclei were first aligned by their static hfs at very low temperatures reached through adiabatic demagnetization.[6] In the latter experiments the polarizations and γ anisotropies obtained are considerably larger than those available by the method proposed here. However, larger activities and longer counting times may be used at the higher temperatures, so that comparable counting statistics may be obtained. The dynamic polarization method yields directly the spin and magnetic moment of the radioactive nucleus and, in fact, may be considered as a microscopic or quantum detector of magnetic resonance. As such, it is unique in that the signal is proportional to the number of decays instead of to the number of atoms as in magnetic resonance absorption, which is a macroscopic detector by contrast. Thus the polarization method is particularly appropriate for the determination of the hfs of short-lived nuclei. That the scheme proposed above will actually work has been demonstrated for 5.3-yr Co^{60}, as described in the following Letter,[7] and for 6-day Mn^{52}.[8]

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† This work was reported earlier at the annual meeting of The American Physical Society in New York, January 30, 1957 [C. D. Jeffries, Bull. Am. Phys. Soc. Ser. II, 2, 65 (1957)].

[1] A. Overhauser, Phys. Rev. **89**, 689 (1953); **92**, 411 (1953).

[2] F. Bloch, Phys. Rev. **93**, 944 (1954); A. Overhauser, Phys. Rev. **94**, 768 (1954); J. Korringa, Phys. Rev. **94**, 1388 (1954); C. Kittel, Phys. Rev. **95**, 589 (1954); P. Brovetto and G. Cini, Nuovo cimento **11**, 618 (1954); P. Brovetto and S. Ferroni, Nuovo cimento **12**, 90 (1954); A. Abragam, Phys. Rev. **98**, 1729 (1955); A. Abragam, Compt. rend. **242**, 1720 (1956); A. Abragam and J. Combrisson, Compt. rend. **243**, 576 (1956); G. Feher, Phys. Rev. **103**, 500 (1956); G. Feher and E. A. Gere, Phys. Rev. **103**, 501 (1956); Pines, Bardeen, and Slichter, Phys. Rev. (to be published); F. M. Pipkin, Bull. Am. Phys. Soc. Ser. II, **2**, 65 (1957).

[3] R. V. Pound, Phys. Rev. **79**, 685 (1950).

[4] A. Abragam, Phys. Rev. **98**, 1729 (1955).

[5] A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 173 (1951); **A206**, 173 (1952); B. Bleaney and D. J. E. Ingram, Proc. Roy. Soc. (London) **A208**, 143 (1952).

[6] See, e.g., Blin-Stoyle, Grace, and Halban, in *Progress in Nuclear Physics* (Butterworths-Springer, London, 1953), Vol. 3, p. 63.

[7] Abraham, Kedzie, and Jeffries, following letter [Phys. Rev. **106**, 165 (1957)].

[8] Abraham, Kedzie, and Jeffries (to be published).

γ-Ray Anisotropy of Co^{60} Nuclei Polarized by Paramagnetic Resonance Saturation*†

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THE proposal of Jeffries[1] in the preceding letter for the polarization of nuclei by the saturation of certain forbidden transitions in paramagnetic resonance has been successfully applied to 5.3-yr Co^{60} contained in a single crystal of $La_2Mg_3(NO_3)_{12}\cdot 24D_2O$.

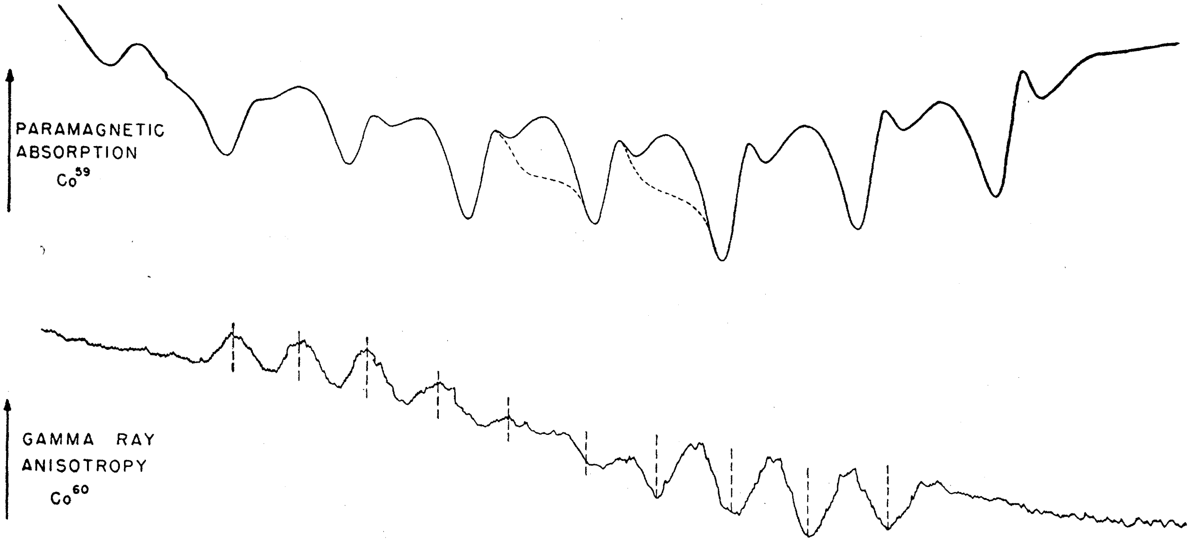

FIG. 1. Paramagnetic absorption and γ-ray anisotropy *vs* magnetic field.

Approximately 1 mC of activity was used, the abundance ratios being Mg:Co^{59}:Co^{60}$=10^4$:50:1. Co^{++} forms two unequivalent magnetic complexes in this crystal,[2] the first having a very small value of B. The second complex has $B\cong 60$ gauss, $A\cong 50$ gauss for Co^{60} and is suitable for the present experiment. The crystal was placed in the microwave cavity of a low-temperature paramagnetic resonance apparatus operating at 9300 Mc/sec. The z axis of the crystal was oriented perpendicular to the dc magnetic field, the microwave magnetic field being approximately parallel to the dc field. Two NaI(Tl) crystals connected via light pipes to photomultiplier tubes were placed just outside the Dewars in directions parallel and perpendicular to the dc field, respectively. The fractional difference in γ counting rate, i.e., the anisotropy ϵ, was recorded on an automatic recorder running at the same speed as the recorder for the paramagnetic resonance absorption. At a constant temperature of 1.6°K obtained by pumping on liquid He, the dc field was slowly varied through the region of the resonance lines with the results shown in Fig. 1. The top trace is the usual modulation derivative of the paramagnetic resonance absorption hfs of stable Co^{59}, observed simultaneously with the γ-ray anisotropy spectrum of Co^{60}, shown below on the same magnetic field scale. The Co^{60} paramagnetic absorption is too weak to be seen. The Co^{59} spectrum shows $2I+1$ main absorption lines corresponding to the known nuclear spin $I=\frac{7}{2}$. The broad lines in between these are the forbidden absorption lines distorted by modulation and power saturation effects. The γ-ray anisotropy spectrum of Co^{60} shows $2I$ lines as expected from the known spin $I=5$. The vertical dotted lines show the positions of the anisotropy lines as calculated from the previously measured[3] magnetic moment ratio $\mu(\text{Co}^{59})/\mu(\text{Co}^{60})$ if one assumes that the nuclear polarization comes from the saturation of the forbidden Co^{60} transitions as discussed in I. From the known decay scheme of Co^{60} and the population distribution among the M states obtained from the simple arguments of I, one can calculate the anisotropy expected, using, e.g., the formulation of Steenberg.[4] The sign of the calculated anisotropy is in agreement with that observed, as is also its reversal about the center point. The relative intensities of the observed anisotropy lines are approximately in agreement with those calculated: 83, 100, 83, 52, 18. The anisotropy observed for the strongest line in Fig. 1 is 1% and is about half that expected for a complete saturation of the forbidden transitions. This indicates incomplete saturation or, more likely, the existence of competing relaxation transitions between adjacent M states.

If the microwave magnetic field is oriented approximately perpendicular to the dc field, weak γ-anisotropy lines corresponding to saturation of the strongly allowed dipole transitions are observed, superposed on the spectrum described above. Under certain conditions there is also evidence of some coupling between the electron $+$Co^{59} system and the electron $+$Co^{60} system, in the sense that saturation of levels in the first system produces population changes in the second system. Full details on this experiment will be published later.

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† This work was reported earlier at the annual meeting of the American Physical Society in New York, January 30, 1957 [Bull. Am. Phys. Soc. Ser. II, 2, 65 (1957)].

[1] C. D. Jeffries, preceding letter [Phys. Rev. **106**, 164 (1957)]; hereafter referred to as I.

[2] R. S. Trenam, Proc. Phys. Soc. (London) **A66**, 118 (1953).

[3] Dobrowolski, Jones, and Jeffries, Phys. Rev. **101**, 1001 (1956).

[4] N. R. Steenberg, Proc. Phys. Soc. (London) **A65**, 791 (1952).