

vector direction of the base field coincides with that of the supplementary rf field; hence the marking intervals become twice as long.

The use of lower rf power, more stable conditions in the excitation of the field, and an investigation of the possibilities of sharp reduction of the rf field after the flight of the particle may all reduce the requirements on stability of the rf amplitude and increase the effectiveness of the marking process.

In addition to synchronous operation with a pulsed accelerator, detection of cosmic particles using triggered rf field trains, and controlled operation of a cloud chamber, it is interesting to consider the possibilities of triggering the rf field from a radiation precursor of a cosmic particle or an early particle in an avalanche of cosmic particles.

<sup>1</sup>D. Gabor and B. Hampton, *Nature* **180**, 746 (1957).

<sup>2</sup>D. Posin, *Phys. Rev.* **73**, 496 (1948).

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## RELAXATION OF DEUTERIUM NUCLEI IN PARAMAGNETIC SOLUTIONS

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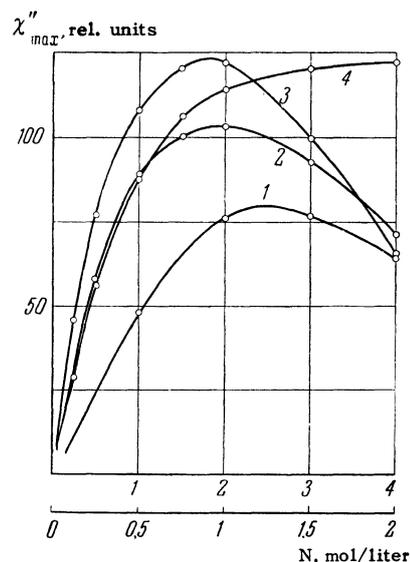
AN investigation has been made of spin relaxation of deuterium nuclei in solutions containing  $\text{Cr}^{+++}$ ,  $\text{Mn}^{++}$ ,  $\text{Fe}^{+++}$  and  $\text{Cu}^{++}$  in heavy water. The measurements were made by means of a modified saturation-curve method<sup>1,2</sup> in which the degree of nuclear saturation is varied by changing the concentration of paramagnetic ions in the solution rather than by changing the amplitude of  $H_1$ , the oscillating magnetic field.<sup>3</sup> All the experiments were carried out at room temperature at  $\nu_0 \cong 2.6$  mc/sec. An analysis of the results indicates the following.

1. The nuclear relaxation time for the same concentration of a given paramagnetic salt in  $\text{D}_2\text{O}$  and  $\text{H}_2\text{O}$  obeys the relation

$$T_{1d}/T_{1p} = (\gamma_p^2/\gamma_d^2)\alpha, \quad (1)$$

where  $T_{1d}$  is the longitudinal relaxation time for deuterons in a  $\text{D}_2\text{O}$  solution,  $T_{1p}$  is the longitudinal relaxation time for protons in a  $\text{H}_2\text{O}$  solution,  $\gamma_p$  and  $\gamma_d$  are the nuclear gyromagnetic ratios ( $\gamma_p^2/\gamma_d^2 \cong 42.4$ ) and  $\alpha$  is a numerical factor. In solutions containing  $\text{Cr}^{+++}$ ,  $\text{Fe}^{+++}$  and  $\text{Cu}^{++}$   $\alpha \sim 4.2$ , with sizable departures toward lower values for copper.  $\text{Mn}^{++}$  ions have much less effect on relaxation of deuterium nuclei:  $\alpha \sim 6.8$  in solutions of  $\text{MnCl}_2$  and  $\text{Mn}(\text{NO}_3)_2$ .

2. As has been indicated earlier in the case of proton relaxation,<sup>2,4</sup> the time  $T_1$  for deuterons becomes much longer (up to a change of one order of magnitude) as a consequence of the production of complexes in the solution in which the water molecules in the hydrate shells of the paramagnetic ions are replaced by other diamagnetic particles. It may be noted, moreover, that the manganese complex behaves the same as the other ion complexes\*  $\text{Cr EDTA}^-$ ,  $\text{Mn EDTA}^{--}$ , and  $\text{Fe EDTA}^-$ , in Eq. (1)  $\alpha \sim 4.2$ .



Curves obtained with solutions of nitrate salts: 1)  $\text{Cu}^{++}$ ; 2)  $\text{Cr}^{+++}$ ; 3)  $\text{Fe}^{+++}$ ; 4)  $\text{Mn}^{++}$ . The upper scale on the abscissa axis refers to Curve 1; the lower scale refers to Curves 2, 3 and 4.

3. In the Figure is shown the maximum intensity of the deuteron resonance line  $\chi''_{\text{max}}$  as a function of the molar concentration  $N$  of the paramagnetic ion in  $\text{D}_2\text{O}$ . The amplitude of the oscillating field  $H_1$  and the amplitude of the modulating field  $H_m$  are fixed. As the quantity  $N$  increases the relaxation time  $T_1$  becomes shorter and the line intensity increases, reaching a value which applies in the absence of saturation. However

this is the case only as long as the line width remains constant, being determined by the inhomogeneity of the field. With further increases in  $N$  the line width goes beyond the limits imposed by the inhomogeneity of the field, causing a reduction in  $\chi''_{\max}$ . An analysis of the curves using the Bloch equation<sup>6</sup> leads to the conclusion that relaxation of deuterium nuclei in paramagnetic solutions is characterized by the condition  $T_1/T_2 \gg 1$  ( $T_2$  is the transverse relaxation time) where the values of  $T_1/T_2$  for the various ions fall in the following order:  $(T_1/T_2)_{\text{Cu}^{++}} > (T_1/T_2)_{\text{Cr}^{+++}} > (T_1/T_2)_{\text{Fe}^{+++}} > (T_1/T_2)_{\text{Mn}^{++}}$ .

These experimental data indicate that the line broadening is due to the displacement of the spin levels of the nuclei by virtue of the interaction of the quadrupole moment of the deuteron with the gradient of the electric field set up by the paramagnetic ion. In this case  $T_1/T_2$  should be approximately proportional to the ratio  $e^2/\mu^2$  where  $e$  is the charge of the magnetic ion and  $\mu$  the magnetic moment of the ion. This relation is actually observed in the experiments. In descending order the ratio  $e^2/\mu^2$  for the various ions is as follows:

$$(2^2/2^2)_{\text{Cu}^{++}} > (3^2/3.8^2)_{\text{Cr}^{+++}} > (3^2/5.9^2)_{\text{Fe}^{+++}} > (2^2/5.9^2)_{\text{Mn}^{++}}$$

The  $T_1/T_2$  ratios are in exactly the same order (cf. above). If the spin levels of the nuclei are equidistant while the probability for  $\Delta m = \pm 1$  transitions is proportional to  $\gamma^2$ , using the general expression for the relaxation time  $T_1$ ,<sup>7</sup> the expression given in (1) results if we take the value  $\alpha = 2$ . The values found in the present work ( $\alpha \sim 4.2$  for solutions containing  $\text{Cr}^{+++}$ ,  $\text{Fe}^{+++}$  and  $\text{Cu}^{++}$ ) would also seem to be explained by the existence of unresolved quadrupole structure in the deuteron lines.

Since the anomaly associated with manganese solutions ( $\alpha \sim 6.8$ ) is not observed in complex ions, it is probably due to the nature of the hydration of the  $\text{Mn}^{++}$  ions in  $\text{D}_2\text{O}$ . It may be assumed that the bonding of the water molecules in the hydrate shell with the paramagnetic ion is slightly covalent. An exception is the  $\text{Mn}(\text{D}_2\text{O})_x^{++}$  ion in which the binding is purely electrovalent. The absence of a covalent bond tends to reduce the magnetic interaction between the neutral ion and the deuterium nuclei in the first coordination sphere, thus increasing  $T_1$ .

\*EDTA ions are ions of ethylene diamine tetra acetic acid. The EDTA ions form stable complex compounds with a stoichiometric ratio of 1:1 with metal ions; the EDTA particle replaces several water molecules in the internal coordination sphere of the central ion.<sup>5</sup>

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<sup>2</sup>A. I. Rivkind, Zhurn. Neorg. Khimia **2**, 1263 (1957).

<sup>3</sup>Bloembergen, Purcell and Pound, Phys. Rev. **73**, 679 (1948).

<sup>4</sup>A. I. Rivkin, Dokl. Acad. Nauk SSSR **100**, 933 (1955).

<sup>5</sup>K. B. Iatsimirskii, Zav. Laborat. (Industrial Laboratory) **21**, 1149 (1955).

<sup>6</sup>F. Bloch, Phys. Rev. **70**, 460 (1946).

<sup>7</sup>C. J. Gorter, Paramagnetic Relaxation (New York, 1947). Russian translation, 1949, p. 107.

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### ANISOTROPY IN MAGNETIC SUSCEPTIBILITY AND DEPENDENCE OF HEAT CAPACITY ON FIELD DIRECTION IN AN ANTI-FERROMAGNET

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It is well known that in a magnetic field  $H$  smaller than some threshold value  $H_0$  the spin wave energy spectrum in a uniaxial antiferromagnet exhibits a marked anisotropy.<sup>1</sup> Thus, for example, if the characteristic antiferromagnetism direction is along the  $z$  axis, in the case  $\mathbf{H} \parallel z$  the energies of the two types of spin waves depend on  $H$  and the wave vector  $\mathbf{k}$  as follows:<sup>2</sup>

$$\varepsilon_k^{(1,2)} = \sqrt{(\mu H_0)^2 + I^2 k^2} \pm \mu H, \quad (1)$$

while in the case  $\mathbf{H} \perp z$ :<sup>3</sup>

$$\varepsilon_k^{(1)} = \sqrt{(\mu H_0)^2 + I^2 k^2}, \quad \varepsilon_k^{(2)} = \sqrt{\mu^2 (H^2 + H_0^2) + I^2 k^2}. \quad (2)$$

Here  $\mu = ge\hbar/2mc$ ,  $H_0 = M_0 \sqrt{J_1(K_1 - K)}$ ,  $M_0$  is the maximum possible magnetization for the given antiferromagnet,  $J_1$ ,  $I$ ,  $K$  and  $K_1$  are the constants in the exchange and anisotropy interactions.\* It is characteristic that in these two cases the ground states of the antiferromagnet are also considerably different: in the case  $\mathbf{H} \parallel z$  the antifer-