

*DYNAMIC POLARIZATION OF PROTONS IN A ROTATING LANTHANUM-MAGNESIUM  
NITRATE CRYSTAL*

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A new possibility of polarization of nuclei in anisotropic crystals by rotating them in a stationary magnetic field and a weak radio-frequency field is reported. A proton polarization amplification coefficient of about 70 has been obtained in a field of 5 kOe and frequency of 165 Mc. Advantages of the method are very liberal magnetic field uniformity and stability tolerances and the use of radio frequencies in the meter range instead of ultra high frequencies.

**I**N the language of spin temperatures, a high degree of nuclear polarization signifies the attainment, in some manner, of a very low nuclear temperature in a relatively large magnetic field.

Recently Abragam<sup>[1]</sup> and Jeffries<sup>[2]</sup> proposed several new methods of lowering the nuclear spin temperature and indicated, in particular, a very simple method suitable for crystals with sharply anisotropic magnetic properties. If such a crystal is placed in a rotating magnetic field  $H_0$ , then the Zeeman energy  $\Delta\epsilon = g(\varphi)\beta H_0$  will oscillate at double the rotation frequency ( $g(\varphi)$  — splitting factor,  $\beta$  — Bohr magneton,  $\varphi$  — angle between the crystal axis and the direction of the magnetic field). If the time  $T_{1e}$  of electron spin-lattice relaxation is comparable with or larger than the period of the revolution, then there is no time to establish equilibrium with the lattice, whose temperature is that of the heat bath  $T_0$ . The electron spin temperature  $T_e$  will therefore vary periodically in approximate proportion to  $\Delta\epsilon$ . In addition, since the probability  $W = 1/T_{1e}$  of the relaxation transitions is proportional to  $g^2(\varphi)$ , the thermal coupling with the lattice breaks as  $g \rightarrow 0$  and the electron spin temperature turns out to be equal to  $T_{e \min} \approx T_0 g_{\min}/g_{\max}$ .

We now consider the behavior of a system of nuclear spins when the nuclear spin-lattice relaxation time is  $T_{1n} \gg T_{1e}$ , as is usually the case. Inside the spin system, however, the equilibrium is reached rapidly via spin-spin relaxation, which does not depend on the temperature. Consequently, if some part of the system is rapidly cooled, then the entire system will first be rapidly cooled to some average temperature, and then the entire

system will slowly return to the initial equilibrium state.

If we assume that the electron and nuclear Larmor precession frequencies  $\nu_e$  and  $\nu_n$  coincide, for example, at an electron spin temperature  $T_{e \min}$ , then both systems become identical and rapid equalization of the spin temperatures should take place via spin-spin relaxation. After many periods of revolution, the spin temperature of the nuclei drops to  $T_{e \min}$ , and consequently the nuclei will acquire the electron polarization.

Robinson<sup>[3]</sup> checked this method and found that in weak fields (1–2 kOe) an approximate tenfold increase in the polarization of the protons actually takes place in a lanthanum-magnesium nitrate crystal  $\text{La}_2\text{Mg}_3(\text{MO}_3)_{12} \cdot 24\text{H}_2\text{O}$  with 2% cerium added. The effect practically vanishes, however, even in a  $\sim 3$  kOe field.

We repeated Robinson's experiment and obtained approximately the same results. The reason for such a slight coefficient of amplification and its sharp decrease with increasing field can be assumed to be the insufficiently strong anisotropy of the crystal ( $g_{\max} = 1.83$ ,  $g_{\min} \approx 0.025$ ). Therefore the magnetic splitting of the levels of the electronic and nuclear spin systems differ greatly for any crystal orientation, and the effect is due only to the finite width of the magnetic levels of the electron system.

Under these conditions, it is meaningful to employ the method of induced transitions, as in the usual effet-solide (see, for example, <sup>[4]</sup>), by acting on the system with a sufficiently strong radio-frequency field. The departure from the usual effet-solide will consist in the time-separation

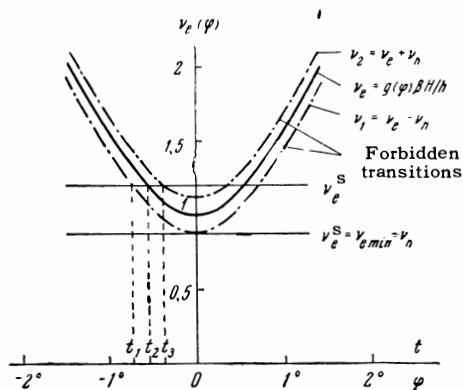


FIG. 1. Dependence of the frequencies of the allowed and "forbidden" transitions on the angle of orientation of the crystal in the magnetic field.

of the relaxation processes and the induced transitions. Second, only positive polarization should be observed, since the forbidden transition with minimum frequency will always be the first to be saturated as the crystal rotates (see Fig. 1). A third difference is that the saturation can take place in principle at any frequency of the radio-frequency field  $\nu_e^S \geq \nu_{e \min} - \nu_n$ . All that changes is the angle between the crystal axis and the direction of the magnetic field at which saturation of the particular transition takes place.

It can be shown that if all three transitions do not overlap, if the intensity of the radio-frequency field is sufficient to saturate the transitions during the time of passage through resonance, and if there is no inversion of the levels of the system on going rapidly through the allowed transition  $\nu_e = g(\varphi)\beta H/h$ , then the amplification will be seven times larger when  $\nu_e^S = \nu_{e \min} - \nu_n$  than when  $\nu_e^S > \nu_{e \min} + \nu_n$ . In the case of level inversion and rapid adiabatic passage through the allowed transition, the amplification does not depend on the saturation frequency.

We verified this method with single-crystal  $(La, Ce)_2Mg_3(NO_3)_{12} \cdot 24H_2O$  and obtained an appreciable increase in polarization. The experiments were made in fields from 2 to 6 kOe at saturation frequencies from 60 to 170 Mc with the crystal rotating uniformly at 30–600 rpm. The experiments were made at 1.3°K.

We established the following:

1. The amplification coefficient increases as the cerium concentration decreases from 2 to 0.2%, namely:

Cerium concentration, %	2	0.8	0.5	0.2
Amplification:	15	25	40	70

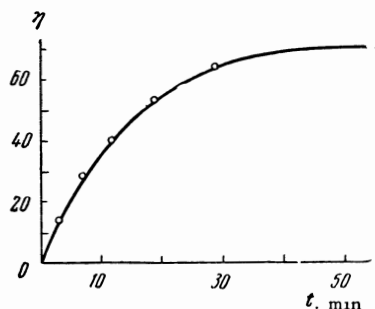


FIG. 2. Amplification of proton polarization vs. the time of saturation of a crystal with 0.2%  $Ce^{3+}$  at frequency  $\nu_e^S = 165$  Mcs in a 5 kOe field.

In the remaining experiments the amplification reached 70 upon saturation within 30–40 minutes (Fig. 2). This value is apparently not the limit and can be increased by a more careful choice of all the parameters.

2. The amplification coefficient increases rapidly as the rotation speed  $f$  increases from 30 to 100 rpm, and then decreases slowly, following approximately the variation of  $T_{e \min}(f)$ .

3. At a power input  $\sim 1$  W and a circuit  $Q \sim 16$ , the intensity of the radio-frequency field was insufficient to produce saturation. To increase the field intensity further it is therefore advisable, besides increasing the  $Q$ , to use a pulsed synchronous supply for the resonant circuit, so as to avoid rapid evaporation of the helium.

4. Only positive polarization takes place at all saturation frequencies. Figure 3 shows the dependence of the amplification coefficient on the magnetic field intensity for a crystal with cerium concentration 0.2% at a saturation frequency  $\nu_e^S = 165$  Mcs. The position of the weak maximum at  $H = 5$  kOe corresponds to  $g \approx 0.024$ , which coincides with the value of  $g_{||}$  indicated in [3].

The presence of a maximum at  $\nu_e^S = \nu_{e \min} - \nu_n$  was predicted above in the case when there is no inversion of the levels and there is rapid passage



FIG. 3. Amplification of proton polarization vs. magnetic field intensity in a crystal with 0.2%  $Ce^{3+}$  at a saturation frequency 165 Mcs at a rotation speed  $f = 250$  rpm.

through the allowed resonance. Inaccurate setting of the crystals and the presence of vibration during the rotation, and also the incomplete resolution and insufficient saturation of the transitions, should lead to a strong decrease and to a smearing of the maximum.

An error in the setting of the crystal can lead to the dependence shown in Fig. 3 also when there is no inversion of the levels of the system. Indeed, if the crystal axis makes an angle  $90^\circ - \varphi$  with the axis of the rotation, and the axis of rotation makes an angle  $90^\circ - \psi$  with the direction of the field, then there are two minimal frequencies during each revolution:

$$\nu'_{e \min} = \frac{\beta g_{\min}}{h} H \left[ 1 + \frac{1}{2} \left( \frac{g_{\max}}{g_{\min}} \right)^2 (\varphi - \psi)^2 \right],$$

$$\nu''_{e \min} = \frac{\beta g_{\min}}{h} H \left[ 1 + \frac{1}{2} \left( \frac{g_{\max}}{g_{\min}} \right)^2 (\varphi + \psi)^2 \right].$$

The presence of two such minimal frequencies can lead to the appearance of a steplike dependence of the amplification coefficient on the field at a fixed saturation frequency.

In conclusion it must be noted that an advantage of this method is the extremely low requirement with respect to homogeneity and stability of the magnetic field, and the use of radio frequencies in the meter band in place of microwave frequencies.

<sup>1</sup>A. Abragam, *Cryogenics* **3**, 42 (1963).

<sup>2</sup>C. D. Jeffries, *ibid.* **3**, 41 (1963).

<sup>3</sup>F. N. H. Robinson, *Phys. Lett.* **4**, 180 (1963).

<sup>4</sup>O. S. Leifson and C. D. Jeffries, *Phys. Rev.* **122**, 1781 (1961).

Translated by J. G. Adashko