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High Proton Polarization Achieved with a (Yb, Y) $(C_2H_5SO_4)_3 \cdot 9H_2O$ Spin Refrigerator in a Nonuniform Magnetic Field

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Proton polarization of approximately 65% has been achieved with the University of Massachusetts "spin refrigerator" in a nonuniform 1.07-T field at 1.25 K. The free protons in (Yb, Y)$(C_2H_5SO_4)_3 \cdot 9H_2O$ are polarized through sample rotation at 100–200 rps. Results represent the first confirmation of high polarization predicted by a single-Yb-ion interaction model since the original proposals of Jeffries and Abragam. The polarized-proton target, with length equivalent to 9 cm of liquid hydrogen, will be utilized in studies of strange particles.

Jeffries and Abragam suggested in 1963 that nuclei could be polarized simply by rotating a suitable crystal in a magnetic field at low temperature.[1,2] The proposed technique involves cyclically transferring a large paramagnetic-ion polarization to nuclear (proton) spins without the use of microwaves and with no constraint on the uniformity of the magnetic field. It requires a very anisotropic g factor for the paramagnetic-ion impurity.

The present experimental results demonstrate conclusively that the early predictions of high proton polarization are realized with ytterbium at $\lesssim 0.04$-at.% concentration in $Y(C_2H_5SO_4)_3 \cdot 9H_2O$. The failure of rather extensive prior efforts[3-7] to achieve high polarizations was *not* due to any fundamental limitation, but rather due primarily to poorly understood effects of higher Yb-ion concentration. Thus, the "spin refrigerator" using ytterbium in yttrium ethylsulfate [$Y(EtSO_4)_3$:Yb] is now very attractive as a polarized proton target.

Langley and Jeffries achieved proton polarizations as large as 19% in 2% Yb-doped $Y(EtSO_4)_3$ at 1.0 T and 1.4 K by rotating a single crystal 2 mm thick by 6 mm in diameter at 60 rps.[3] Subsequently McColl and Jeffries utilized a combination of a pulsed cross field with a dc field to rotate the magnetic field with respect to a fixed $Y(EtSO_4)_3$:Yb crystal.[4] These latter studies achieved proton polarization of 35% in 2% Yb-doped $Y(EtSO_4)_3$ at 1.2 to 1.4 K, with $B_{dc} = 15$ kG and $B_{pulsed} = 20$ kG. (*Note.*—All Yb concentrations refer to those of the growing solution; actual concentrations in crystal samples were approximately half the indicated values.)

As indicated by Jeffries in review papers,[8,9] the successful experiments by Schmugge and Jeffries utilizing the microwave technique to achieve proton polarization of 70% with Nd-doped lanthanum magnesium nitrate (LMN) led to the extensive use of microwave targets and caused most physicists to cease working on spin refrigerators.[10] Nevertheless, Potter and Stapleton at the

University of Illinois carried out the development of a helium-gas-turbine drive for a $Y(EtSO_4)_3$:Yb sample.[5] They achieved 35% proton polarization at 1.7 K, with a 1.5-cm^3 polycrystalline sample of 0.04% Yb-doped $Y(EtSO_4)_3$ rotated at 120 rps in a dc field of 1.13 T. The Potter-Stapleton work established that the dominant cross-relaxation process is a one-to-one spin flip for the Yb ions and neighboring protons, and hence the proton polarization can approach that of the Yb ions. Internal heating of the sample was found small. Potter predicted that proton polarization of 40–60% would be possible. Brom and Huiskamp, employing the McColl-Jeffries technique at lower temperatures, obtained rather low polarization with $Y(EtSO_4)_3$:Yb; but they estimated that proton polarization of more than 70% could be realized under suitable conditions with low Yb concentration.[7]

As elucidated by Jeffries and others, the mechanism of polarizing the free protons of $Y(EtSO_4)_3$:Yb (in both the ethyl and the water groups) depends on the Yb-ion g factor and (less significantly) on its relaxation rate. In the presence of the external magnetic field, the Yb ions populate the lowest of four Kramers doublets. This doublet has an anisotropic g factor, $g(\theta)=(g_{\parallel}^2\cos^2\theta+g_{\perp}^2\sin^2\theta)^{1/2}$, where θ is the angle between the applied field and the crystal c axis. With the effective spin taken as $\frac{1}{2}$, the Yb $g_{\perp}$ is as small as the proton g value of 0.003 04, while $g_{\parallel}=3.33$.[11] The Yb-ion relaxation rate varies approximately as $\sin^2\theta\cos^2\theta$. If the crystal is rotated with respect to the magnetic field at a rate greater than the Yb-ion relaxation rate, then the Yb-ion polarization assumes an almost *constant* value corresponding roughly to $g(45°)$, or $\tanh(2.4\mu_B B/2kT)$. As θ approaches 90°, the energy-level difference of the doublet greatly diminishes so that it nearly matches the equilibrium energy splitting of the protons. At 90°, a dipole-dipole interaction may cause the Yb spin orientation to be exchanged with that of a nearby proton. The proton relaxation rate drops rapidly beyond 90°. The polarization process continues with further rotation: At $\theta=180°$, g^{Yb} is again large; and at $\theta=270°$, $g^{Yb}\approx g^{p}$. "Spin diffusion" among the protons spreads the polarization from "near" to "far" protons.

The (nonequilibrium) polarization enhancement of the protons is equivalent to reducing their temperature by several orders of magnitude; hence, the technique has been described as "spin refrigeration."

The advantages that Jeffries pointed out[9] make the spin-refrigerator target very attractive for use in a bubble chamber, a streamer chamber, or a spark-chamber facility. These advantages are the following: the relatively low cost and ease of operation; the independence from restrictions on field uniformity; the low helium consumption; and (compared with the LMN microwave targets) the relatively large proportion ($\frac{1}{9}$) of polarized to unpolarized (bound) protons. Also, typical buildup time is short and relaxation time is long.

The University of Massachusetts has developed a spin-refrigerator polarized target with the expectation of utilizing it for such processes as $K^-+p\to\Lambda+\pi^++\pi^-$ and $\pi^-+p\to K^0+\Lambda$ or $K^{*0}+\Lambda$. The *polycrystalline* target samples employed have been approximately 1 in. and 3 in. in length, with 1 in. diam. The design of the continuous-flow (Roubeau) type of cryostat and associated pumping systems was based primarily on the work of engineer Ralph Niemann of Argonne National Laboratory, and was intended to reach 1.0–1.2 K with a heat load of approximately 1.5 W. The internals were initially developed for a helium-gas-turbine drive, but this was not very satisfactory. Our final design, therefore, utilizes a motor drive (a "Selsyn" system) with a G-10 shaft coupled rigidly to the target. Nylon ball bearings in Delrin races constrain the shaft. (Even small rotating components must be nonmetallic.) The cryostat may be used in any orientation from near-horizontal to vertical.

Figure 1 presents a cross-sectional view of the cryostat and internals. Liquid helium at 4.2 K is delivered through a transfer line to a "separator tank"; cold gas is pumped through tubing soldered to the conical section, while liquid helium is forced through the helical Parkinson heat exchanger to a needle valve, and thence to the target region. The main exhaust system pulls cold helium gas from the target region past the heat exchanger and through the heat baffles of the conical section; it is provided with a 1300-ft^3/min Roots pump connected to the upper section of the cryostat by 10-in. pipe.

The NMR system which measures polarization involves modulating the magnetic field at 25 Hz and sweeping entirely through the resonance line.[12] It is to be noted that the NMR measurement does impose a constraint on field uniformity; it is desirable that the total variation be less than approximately 3 mT (30 G) within the region seen by the NMR coil. (This is to be contrasted with

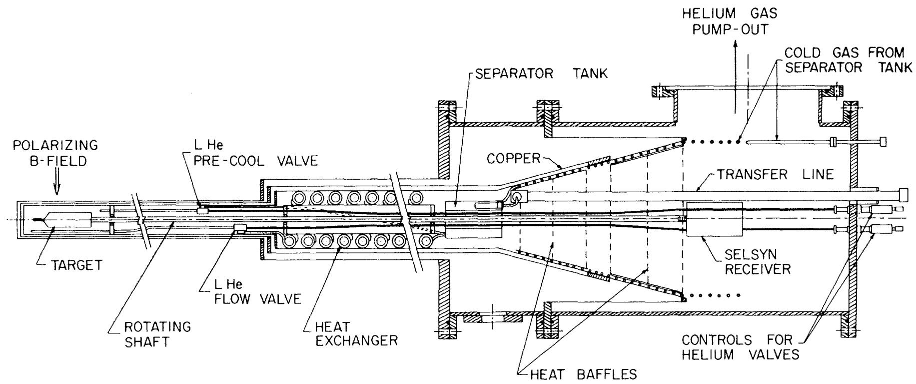

FIG. 1. The University of Massachusetts polarized target. The continuous-flow cryostat, of approximately 60 in. length, may be used in various orientations. Inner and outer jackets are of stainless 304.

the microwave-target limit of 0.5 mT on the field variation throughout the *entire target*.)

Measurements taken in recent months have yielded proton polarizations of at least 65%. Experimental conditions have varied from 1.25 K (1-in. sample) to 1.45 K (3-in. sample) with rotational frequencies from 50 to 200 rps; the magnetic field has been 1.07 T. These *proton* polarizations are close to the calculated steady-state polarization of the paramagnetic Yb ion; they thus indicate that one-to-one spin flips dominate, and that during each cycle the transfer of energy from the protons to ions and on to the lattice is as nearly complete as possible, i.e., $\epsilon = 1$, $K \rightarrow 1$, and $f_\epsilon \rightarrow 1$ in Eqs. (16) and (17) of the second paper of Ref. 5.

Figure 2(a) presents the proton polarization versus rotational frequency for measurements approaching asymptotic values and compares these with calculations of the Yb-ion polarization (and maximum expected proton polarization) for various temperatures. Figure 2(b) shows the calculated ion polarization versus frequency for various magnetic fields. (Calculations are for single crystals. Averaging for a polycrystalline sample could reduce predictions by approximately 10%.)

The buildup of polarization has been found, as expected, to be nearly a pure exponential. The characteristic time depends inversely on Yb-ion concentration ($\approx 0.02\%$ for our crystalline sample); it is found to be about 10 min at 120 rps. Buildup time is observed to vary linearly with rotation frequency; this provides further evidence of complete energy transfer during each cycle.

Figure 3(a) presents the relaxation rate for proton polarization τ_{1n}^{-1} versus temperature, the rate being proportional to concentration. The open circles are for the most recent crystals grown at the University of Massachusetts; they fall close to calculated values with 0.02% Yb concentration assumed. Figure 3(b) presents calculated relaxation rate versus magnetic field with measurements indicated. At 0.2 T the relaxation time apporaches twelve days at 1.0 K. The curves in Fig. 3 are calculated for a single-paramagnetic-center model using parameters from measured ion-spin-lattice relaxation in $Y(EtSO_4)_3$:Yb.[11,13]

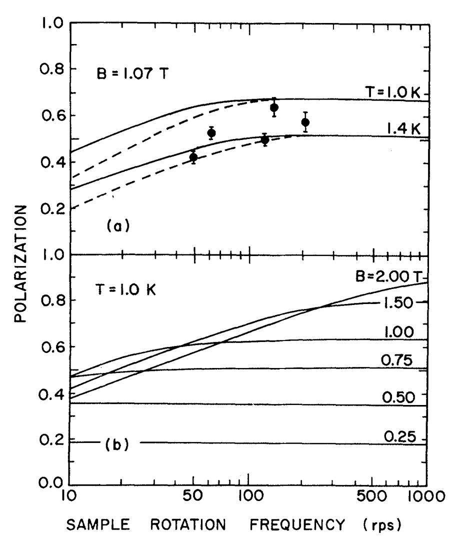

FIG. 2. (a) Measurements of near-asymptotic proton polarization vs rotational frequency. These are compared with calculations of Yb-ion polarization (and maximum expected proton polarization, dashed lines). The 65% polarization at 130 rps was at 1.25 K. The 58% polarization measurement at 200 rps was made with the 3-in. sample at 1.45 K. (b) Calculation of Yb-ion polarization vs frequency at various magnetic fields.

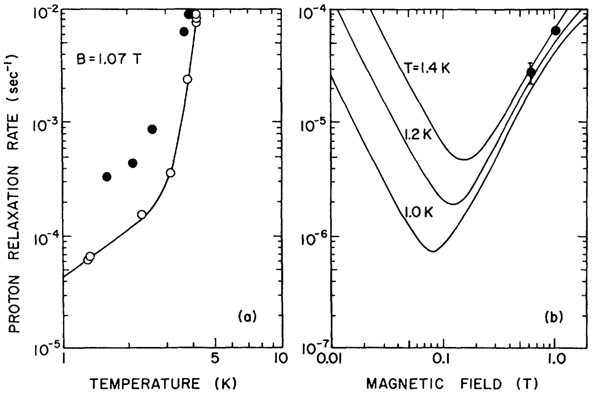

FIG. 3. (a) Proton relaxation rate vs temperature for two samples of $Y(EtSO_4)_3$:Yb, with one set of measurements compared with theoretical results for 0.02% Yb concentration. (b) Calculated proton relaxation rate vs magnetic field. The experimental point at 1.07 T is a University of Massachusetts measurement at 1.3 K; that at 0.6 T was obtained in studies at the University of California, Davis, at 1.1–1.2 K.

A possible configuration for use of the polarized target is to enhance the magnetic field of a spark-chamber facility by use of a modest Helmholtz-coil arrangement during the polarizing operation and then to stop rotation, reduce the total field, and thus capitalize on the longer relaxation time at the lower field.

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Knock-Out Nucleons from Relativistic Nuclear Collisions

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A nonequilibrium single-scattering mechanism is proposed to describe the inclusive proton spectrum from relativistic heavy-ion collisions. Data from ^{20}Ne + U and ^{4}He + U collisions are shown to be consistent with a simple model of the knock-out process. Two-proton azimuthal angle correlations are suggested as a unique signature of this reaction mechanism.

The inclusive proton spectra from relativistic ^{20}Ne + U and ^{4}He + U collisions have been shown to be consistent with the existence of a nuclear "fireball" at temperatures of 30–50 MeV.[1] The composite particle spectra from these collisions[2] also show equilibrium features.[3] However, a literal interpretation of the fireball picture requires equilibration to be achieved on time scales $\lesssim 10^{-22}$