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Test of a new polarized hydrogen target (spin refrigerator) as neutron polarizer

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Neutron-transmission measurements were made to determine the proton polarization in a single crystal of yttrium ethylsulfate with the aim of exploring its utilization as a spin filter for thermal and epithermal neutrons. The proton polarization is coupled to that of a paramagnetic ytterbium impurity simply by rotating the extremely anisotropic crystal in a magnetic field (spin refrigeration). A proton polarization of 41% was obtained in a helium bath near 1.2 K by rotating a large single crystal at a frequency of 150 sec^{-1} in a field of 14.6 kOe. The extrapolation of the results indicates that a proton polarization $p_H \simeq 65\%$ can be achieved with relatively simple technology. For this value of p_H the spin refrigerator becomes a competitive neutron spin filter.

INTRODUCTION

The advent of pulsed neutron sources dedicated to condensed-matter studies has spurred the development of new ways of polarizing thermal and epithermal neutron beams. The knowledge of the neutron-spin state, in addition to that of its energy and momentum, gives the possibility of separating spin-dependent from spin-independent processes in the interaction of neutrons with matter, important both in nuclear physics and in the neutron scattering study of magnetic materials. In a conventional, steady-state neutron source the energy of the incoming beam is to be defined; this is usually obtained in the thermal range by Bragg reflection of a monochromating crystal. Polarization is then required for only one wavelength, and this is accomplished by choosing as monochromator a special magnetized crystal with an extremely spin-dependent reflectivity.[1] If the neutron source is pulsed, the energy of the beam arriving at the sample at a certain instant is defined by its time of flight; hence, all the wavelengths can be utilized. Thus, the need arises for polarizing a "white" neutron beam.

The polarization of the neutron beam can be obtained by using a neutron-spin filter; i.e., a filter that contains a material which effectively removes neutrons of only one spin state from the beam, either by preferential absorption or scattering. It is well recognized[2] that a slab of material containing polarized hydrogen nuclei constitutes a most effective spin filter. The trick is to efficiently polarize the hydrogen nuclei without requiring external conditions and apparatus of such complexity that it would preclude having a neutron-spin filter that could run unattended for a long period of time. The goal of the present work is to describe a new neutron-spin filter, in which high hydrogen polarization is attained by the process called "spin refrigeration," and the tests performed on a prototype.

NEUTRON-SPIN FILTERS

We will briefly review the function and the characteristics of the available or proposed neutron-spin filters. To start with, we introduce the two nuclear cross sections σ^+ and σ^- for neutrons polarized parallel and antiparallel to the polarization of a nucleus of spin I. For σ^+, a pure state of the compound nucleus $(I+\frac{1}{2})$ is formed, while for σ^- a mixed state is formed ($I+\frac{1}{2}$ and $I-\frac{1}{2}$). Suppose that the $I-\frac{1}{2}$ state is absorbing; then a sufficiently thick filter simply removes neutrons of the wrong spin from the beam, and does not attenuate the remaining half of the beam if the nuclei are fully polarized. A filter approaching this condition has been built[3] using a target of polarized ^{149}Sm. However, its best performance is limited to a range of neutron energies close to the resonances of samarium.

Filters based on preferential scattering are capable of polarizing neutrons of a broad energy range; their efficiency is only moderately wavelength dependent as illustrated for the case of hydrogen which has the most favorable ratio σ^+/σ^- of all elements.

For neutron energies high compared with the energy of chemical bonding of the hydrogen, the scattering process can be considered as that of a free particle. For a proton polarization p_H, the cross sections are[2]

$$\sigma^{\pm}=(19.9\mp1.67p_H)\times10^{-24}\ \text{cm}^2\ . \tag{1}$$

At lower energies the effects of the chemical bonding become progressively important. The scattering can be separated into three components, namely incoherent, inelastic, and elastic scattering. The inelastic scattering cross section becomes small for low values of the temperatures of the sample and of the neutron beam. If the target is crystalline (rather than glassy), no diffraction is present for neutron wavelengths longer than the Bragg cutoff. Hence, for low neutron energies (of the order of 1 meV) the cross sections take the form

$$\sigma^{\pm}=\sigma_0(1-\tfrac{1}{3}p_{\mathrm{H}}^2\mp\tfrac{2}{3}p_{\mathrm{H}})\,, \tag{2}$$

with a value of the cross section, extrapolated to zero neutron energy, of $\sigma_0=79.7\times10^{-24}$ cm^2.

The expressions for transmission and polarization of an initially unpolarized neutron beam are

$$T=\tfrac{1}{2}[\exp(-\sigma^{+}nt)+\exp(-\sigma^{-}nt)]\,, \tag{3}$$

$$p_n=\tfrac{1}{2}[\exp(-\sigma^{+}nt)-\exp(-\sigma^{-}nt)]/T\,, \tag{4}$$

where n is the number of protons per cm^3 and t is the target thickness. Figures 1 and 2 present the values of T and p_n as calculated for a 1-cm-thick crystal of yttrium ethlysulfate nonahydrate ($n=5.57\times10^{22}$ protons/cm^3). From these figures several conclusions can be drawn. First, the thickness of the filter has to be tailored to the desired range of neutron energies. Second, the acceptable transmission and polarization depend on the type of experiment to be performed. If we desire a minimum neutron polarization of 95% and a transmission of not less than 7%, we see that the minimum hydrogen polarization should be no less than 60%. Finally, it can be observed that the neutron transmission is very sensitive to the degree of polarization of the protons in a crystal of the proper thickness. Measurements of this kind at either end of the thermal spectrum are easy to perform and interpret, and thus they provide an excellent test of the proton polarization.

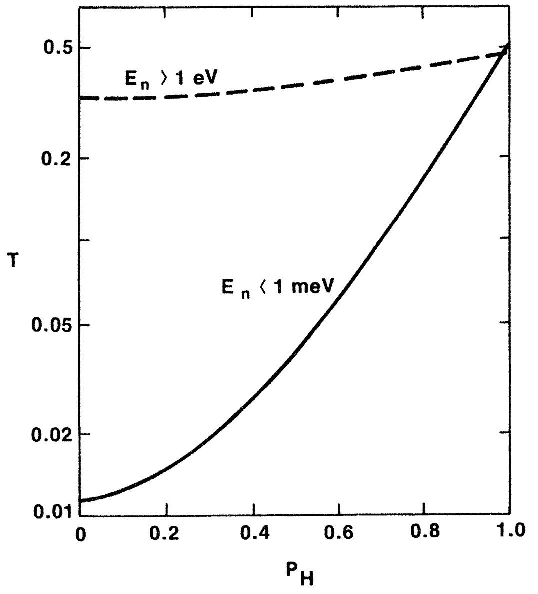

FIG. 1. Transmission of a neutron beam through a polarized target containing 5.57×10^{22} protons/cm^2; neutron energy at both ends of the thermal spectrum.

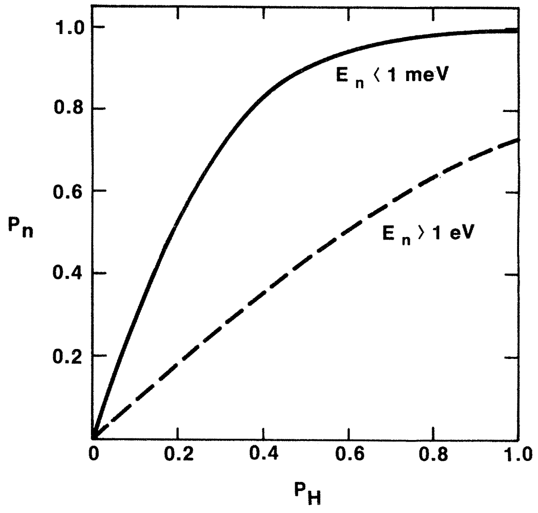

FIG. 2. Polarization of a neutron beam in the same conditions specified in Fig. 1.

In principle, the most direct way to attain sizable hydrogen polarization is by "brute force," namely by placing the target in a high external field (of the order of 10^5 Oe) and cooling it to a temperature of the order of a few mK. Although the technology to achieve such a feat exists, at present the costs of construction and sustained operation of such a filter are unacceptable. The alternative is to polarize the hydrogen dynamically, i.e., by coupling the nuclear spins of the hydrogen with those of a paramagnetic impurity contained in the target. Two methods have been developed to achieve this dynamic polarization transfer: the spin refrigerator and the so-called solid-state effect. The latter is the older and better-known approach to the problem.[4] A diamagnetic, hydrogenated insulator contains a small concentration of fixed paramagnetic impurities of Larmor frequency ω_e. The application to the sample of microwave power on either side of the electron-spin resonance at a frequency $\omega=\omega_e\pm\omega_{\mathrm{H}}$ (where $\omega_{\mathrm{H}}\ll\omega_e$ is the Larmor frequency of the nuclear spin of the hydrogen) can lead to an enhanced polarization p_{H}, which can approach that of the electronic polarization of the impurity p_e. This method has been widely applied to obtain polarized targets in high-energy physics,[5] and in nuclear physics to polarize neutrons in the keV range.[6] Experimental filters of this kind have also been constructed and tested at several neutron sources for studies of solid-state physics.[2,7,8] While a satisfactory proton polarization has been obtained, construction and continuous operation of such filters presents some difficulties. The solid-state effect technique demands a high homogeneity of the applied magnetic field and a high stability of the applied microwave frequency. For the highest proton polarization the temperature must be below 1 K with the target immersed in a pumped ^{3}He bath. Unfortunately, ^{3}He is a strong neutron absorber and thus cannot be placed in the direct neutron beam; consequently the cooling efficiency is greatly reduced.

In the spin refrigerator the high electronic polarization of a paramagnetic impurity, contained in an extremely anisotropic crystal, is transferred to the hydrogen spins simply by rotation. This technique (proposed independently by Abragam[9] and Jeffries[10]) has been examined[11–14] for a number of years, without reaching a competitive proton polarization and without reaching a clear understanding of the factors that limited the polarization. However, a more recent report[15] presented evidence that a proton polarization upward of 65%, as determined by nuclear-magnetic-resonance methods, had been achieved for easily achievable conditions; namely, for a sample temperature of 1.2 K, a rotational speed of 120 $\sec^{-1}$ and an applied magnetic field of 12 kOe. This has made the system seem quite attractive and worthwhile to investigate for use as a neutron-spin filter.

SPIN REFRIGERATOR

At the core of the spin-refrigerator system is a crystal of yttrium ethyl sulfate, $Y(C_2H_5SO_4)_3 \cdot 9H_2O$—YES for short—a hexagonal crystal in which ~0.01 at. % of the yttrium is substituted with paramagnetic Yb^{3+} ions. The Yb^{3+} ions in the crystal field of YES have an effective spin of $\frac{1}{2}$; they constitute a two-level system with a very anisotopic g factor,[12]

$$g(\theta)=(g_{\parallel}^2\cos^2\theta+g_{\perp}^2\sin^2\theta)^{1/2}\ , \tag{5}$$

where $g_{\parallel}=3.33$ and $g_{\perp}$ is of the order of the gyromagnetic ratio of the proton. The Yb^{3+} relaxation time[16] is very anisotropic,

$$1/T_{1e}\simeq AH^4T\sin^2\theta\cos^2\theta\ , \tag{6}$$

with $A=2.16\times10^{-13}$ $\mathrm{Oe^{-4}K^{-1}sec^{-1}}$ (in contrast, the proton spins have energy levels independent of θ). The crystal is rotated in a magnetic field so that the angle θ between H and the hexagonal c axis varies from 0° to 90° to 180°. When $\theta \sim 45°$ the Yb spins become rapidly polarized, and they remain polarized when the crystal is rotated toward $\theta=90°$ (provided this is done quickly compared to T_{1e}, but slowly compared to the Larmor period). In other words, the population of the two Yb spin levels remains unchanged, while the Zeeman splitting shrinks according to $g(\theta)\beta H$ toward the value of the Zeeman splitting of the protons. Close to $\theta=90°$ the Yb spin finds itself able to undergo a mutual spin flip with a neighbor proton through dipole-dipole coupling, with the Yb flipping to the higher-energy state, and the proton to the lower, in the direction of H; the polarization is thus transferred to the proton. The process, illustrated schematically in Fig. 3, is repeated every half-cycle. If the crystal contains N_H protons and N_e Yb^{3+} impurities, after approximately N_H/N_e half-cycles the proton and ytterbium polarizations are in "equilibrium," at the electronic polarization at $\theta=45°$, as if the proton-spin temperature were much lower ($\sim10^{-3}$) than the actual temperature of the thermal bath. Hence, the term "spin refrigerator." The protons are eventually depolarized by relaxation, but at a much slower rate than in the polarization process.

The polarization of the Yb at the cross-over point in conditions of dynamic equilibrium is[12]

$$\bar{p}_{es}=\frac{1-K_e}{K_e}\int_0^{\tau}\exp\left[\int_0^{t}T_{1e}^{-1}(t')dt'\right]\frac{p_{e0}(t)}{T_{1e}(t)}dt\ , \tag{7}$$

where T_{1e} is given by Eq. (6), τ is the time between two cross-relaxation regions, and $p_{e0}(t)$ is the thermal equilibrium electronic polarization for the configuration of the system at the time t. K_e is the probability of restoring the electronic polarization in the time τ after cross relaxation,

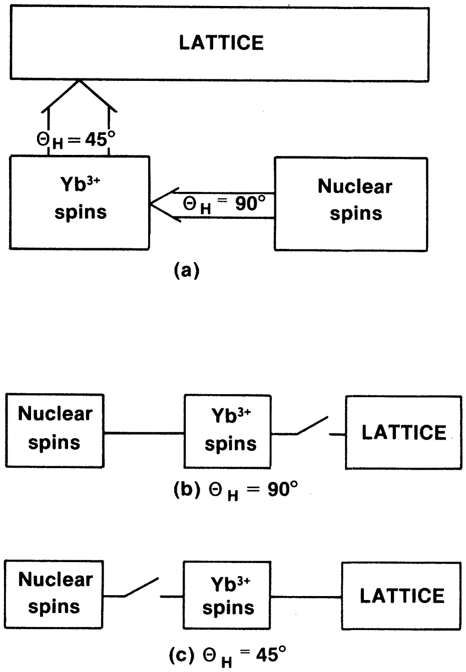

FIG. 3. Scheme of the magnetic coupling in YES as a function of the angle between the applied magnetic field and the c axis of the crystal.

$$K_e=1-\exp\left[-\int_0^{\tau}T_{1e}^{-1}(t)dt\right]\ , \tag{8}$$

and is a function which rapidly saturates as the magnetic field is increased.

In Fig. 4 we present plots of $\bar{p}_{es}(H)$ for values of the temperature and the rotation frequency in the range of the experimental observations. For low fields, $\bar{p}_{es}\propto H/T$, independent of the rotation frequency, which instead becomes important close to the saturating magnetic field values. In any case, to achieve a proton polarization greater than 60%, the transfer of polarization from the

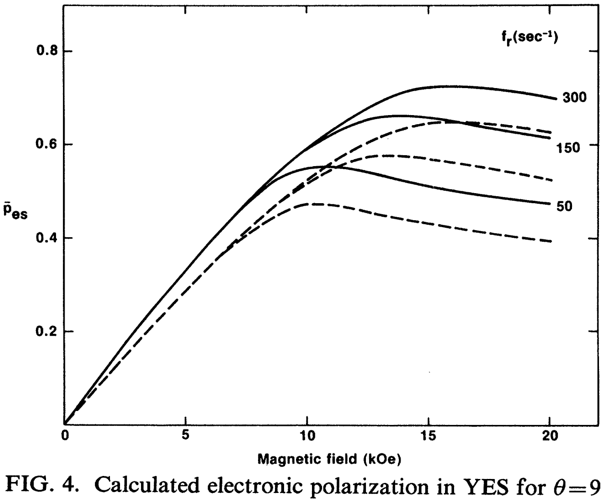

FIG. 4. Calculated electronic polarization in YES for $\theta=90°$. Continuous line: $T=1.1$ K. Dotted line: $T=1.3$ K.

electronic to the proton system must be almost complete.

The proton polarization—and its rise time—may be written in the approximate form[12]

$$p_{\mathrm{H}}=\frac{K_e\bar{p}_{es}\langle \epsilon f_\epsilon\rangle}{\langle \epsilon^2 f_\epsilon\rangle-(1-K_e)\langle \epsilon f_\epsilon\rangle^2}\,, \tag{9}$$

$$\frac{1}{\tau_0}=\frac{1}{\tau}\frac{N_e}{N_{\mathrm{H}}}[\langle \epsilon^2 f_\epsilon\rangle-(1-K_e)\langle \epsilon f_\epsilon\rangle^2]\,, \tag{10}$$

which are written assuming that the nuclear relaxation time is much longer than τ_0 and that the thermal equilibrium proton polarization is very small. The terms $\langle \epsilon f_\epsilon\rangle$, etc. represent the coupling between the electron- and nuclear-spin systems averaged over the spin-flip efficiencies of the different protons, and also for the multiple-spin processes. These come about for the following reason. One proton spin flips with one ytterbium (1:1 process) only when $g(\theta)\leq g_n$. However, for $g(\theta)\leq 2g_n$, two protons can flip with one electron (2:1 process). These terms are explicitly included in Eqs. (9) and (10), where $\epsilon=1$, 2, and 3 for the 1:1, 2:1, and 3:1 processes. Note that the presence of multiple spin flips would degrade the performance of the spin refrigerators; for example, if 2:1 processes were predominant, the maximum proton polarization achievable is only half that of the electronic polarization. The detailed evaluation of the flipping efficiency f_ϵ has been extensively reported in the literature.[11–14] Unfortunately, there is still much uncertainty surrounding the calculation of the $\langle \epsilon f_\epsilon\rangle$. For our purpose it is sufficient to treat the $\langle \epsilon f_\epsilon\rangle$ as empirical parameters, and to examine the conditions under which the coupling parameter between p_{H} and $\bar{p}_{es}$ can be brought close to unity.

GROWTH OF YES CRYSTALS

In a target made of an aggregate of randomly oriented crystallites—rather than a single YES crystal—it is still possible to achieve high proton polarization, since for each of the granules the rotating magnetic field crosses the hexagonal plane at some time during the cycle. However, the averaging of the orientations causes some degradation of the performance, making the use of single crystal preferable.

As a neutron-spin filter it is also preferable to have a single-crystal target. In the thermal neutron range, the probability of unwanted Bragg scattering is much higher for a powder. In the second place, the coarse-grained samples that have been extensively used in previous NMR experiments offer a highly inhomogeneous cross section to the neutron beam, making the choice of an optimal thickness impossible.

For a practical neutron application, the lateral size of the crystal should match the opening of the neutron beam, typically 5—10 cm^2. Thus some effort was spent in the growth of sizeable single crystals. Single crystals of pure and Yb-doped YES were grown from aqueous solution in a rotary crystallizer. YES was formed by reacting a slurry of $Y_2(SO_4)_3$ with a solution of barium ethylsulfate (BES). The yttrium sulfate was prepared from high-purity Y_2O_3 and H_2SO_4. For the growth of Yb-doped crystals, Yb_2O_3 (enriched to 97.15 at.% in ^{172}Yb) was added to the Y_2O_3 prior to synthesis of the sulfate. The $Y_2(SO_4)_3$-BES mixture was stirred for about 1 d at room temperature to complete the reaction. The resulting $BaSO_4$ was removed by filtration and the solution was evaporated under dry N_2 at room temperature until saturated.

The overall development of the growing crystal depends on many parameters, including the mass deposition rate, rotation rate, and orientation of the crystal with respect to the rotation axis. For "pure" YES solutions, it was not possible to grow sizable crystals under any conditions. Growth occurred only along the hexagonal *c* axis leading to the development of long, hexagonal prisms. The growth normal to the *c* axis is enhanced, however, by adding excess BES. Approximately equidimensional crystals could be grown from a solution containing 0.01 to 0.02 g Ba/cm^3. The maximum [Ba]/[Y] ratio in the resulting crystals was 1.3×10^{-4}.

The rotary crystallizer contained 2 l of saturated solution, and two crystals were grown at the same time. The solution was seeded at 286 K. After a slow initial growth (to 1—2 cm^3 crystal size), the solution temperature was lowered by 0.15 K per day to yield a mass deposition rate of 1.2 g per day. The best crystals (quite transparent, with a minimum amount of solution inclusions) were grown with an average lineal velocity of 5 cm sec^{-1} (10 rpm reversed every 5 min) and with the hexagonal *c* axis parallel to the rotation axis. Hexagonal prisms [bounded by (10.0) planes and capped by (11.1) and (10.1) facets] up to 60 g (34 cm^3) were grown.

On the basis of the past experience[15] the content of ytterbium was kept very low in order to minimize direct magnetic interactions between the impurity ions. Crystals were grown from two different Yb-doped solutions. The [Yb]/[Y] atom ratio in the grown crystals was 0.9×10^{-4} and 2.6×10^{-4}—as determined by flame spectroscopy—and the effective distribution coefficient $([\mathrm{Yb}]/[\mathrm{Y}])_{\mathrm{solid}}/([\mathrm{Yb}]/[\mathrm{Y}])_{\mathrm{solution}}$ was 0.19 and 0.16, respectively. The addition of Yb to the solution tended to reduce the growth normal to the *c* axis. Indeed, it may not be possible to obtain equidimensional crystals with a Yb concentration much greater than our highest value.

Samples of pure YES were used for an analysis of the crystalline properties of the material. In a neutron-diffraction study[17] the positions of the hydrogen atoms were determined, and the overall symmetry—previously determined by x-ray structural analysis[18]—was confirmed. YES crystallizes in the hexagonal structure with space group $P6_3/m$, with cell parameters $a=13.871$ Å and $c=7.007$ Å at room temperature.

NMR EXPERIMENTS

The proton polarization of the new single crystals of YES:Yb was initially tested by NMR. In this technique the enhancement of the proton resonance line with the sample situated in a rotating magnetic field is measured in comparison with that at thermal equilibrium. The enhancement is directly proportional to the difference of the population levels of the two $I=\pm\frac{1}{2}$ states.

For the experiments, conducted at the University of

California at Davis, two techniques were used. In the first a steady magnetic field was applied perpendicular to the c axis of the YES crystal, while an oscillating magnetic field was parallel to the c axis. In the second apparatus, an electrically rotating magnetic field was applied in the a-c plane. In both cases, extreme measures were taken to minimize the possibility of systematic errors in the measurement and calibration of the greatly enhanced NMR signals which can lead to erroneous determinations of the proton polarization. Also, because the sample was stationary, the temperature of the ^{4}He bath and thus the sample temperature could be reliably determined.

In Table I we present a summary of the measurements performed in an oscillating magnetic field. The proton polarization is considerably lower than the calculated electronic polarization in these relatively low magnetic fields, but increases for the higher-field values within the set. The approach to saturation followed an exponential function quite rigorously up to $3\tau_0$. The value of $\tau N_{\mathrm{H}}/N_e$ [cf. Eq. (10)] for the two ytterbium concentrations used is, respectively, 51 min (for $[\mathrm{Yb}]/[\mathrm{Y}]=0.9\times10^{-4}$) and 18 min (for $[\mathrm{Yb}]/[\mathrm{Y}]=2.6\times10^{-4}$) or considerably longer than the experimental τ_0, again indicating incomplete coupling. Perhaps the most direct indication on the behavior of the sample comes from an estimate of the effective number of the protons that flip for each Yb spin flips, defined as $\epsilon_{\mathrm{eff}}=\langle\epsilon^2 f\rangle/\langle\epsilon f\rangle$. The results, as obtained from the experimental data with the aid of Eqs. (9) and (10), indicate that at low fields multiple spin flips are quite important, but also that their weight decreases rapidly by raising the magnetic field.

NMR measurements were also made to obtain information on the gyromagnetic ratio of the Yb^{3+} spins normal to the c axis. Ideally, a perfect C_{3h} point symmetry of the Yb^{3+} ion in the YES crystal with its single unpaired $4f^{13}$ electron would lead to a complete vanishing of the components of $g_\perp$. There are, however, a number of mechanisms that can admix the ground and excited states and explain the small yet nonzero value of $g_\perp$. One of these is a third-order Zeeman splitting which becomes important in high magnetic fields. This gives rise to a term of the form

$$g_\perp=g_{\perp0}+g_{\perp6}\cos6\phi\ , \tag{11}$$

where $g_{\perp6}$ is proportional to H^2, and ϕ is the turn angle in the hexagonal plane. While the detailed field dependence of $g_{\perp0}$ on the magnetic field cannot be assessed directly, the importance of $g_{\perp6}$ can be verified by measuring the proton polarization with fields oscillating in the a-c plane and the b-c plane of the single crystal. A series of measurements with an oscillating field of 6.3 kOe ($H_{\mathrm{dc}}=3$ kOe) showed that the proton polarization is completely independent on the ϕ angle; hence, such a term is considered to be unimportant.

Finally, the NMR measurements shed some light on the problem of the nonrelaxation heating, which comes about because in a rotating field the Yb polarization does not relax sufficiently fast to be in phase with its value at thermal equilibrium. Hence, the spin system absorbs energy from the agent rotating the crystal (or from the oscillating magnetic field), and in turn this energy is transferred to the lattice by spin-lattice relaxation. The heating might be more severe for sizable single crystals in view of the thermal resistance at the surface or Kapitza resistance; in other words, the temperature of the sample might be higher than the reference temperature of the cooling helium bath. However, as it appears from Table

TABLE I. Nuclear-magnetic-resonance measurements in an oscillating field H_{osc} parallel to the c axis, with steady field H_{dc} normal to it. ($f_r=60$ sec^{-1}; $T=1.2$ K.)

Sample size (mm) $a\times b\times c$ axes	[Yb]/[Y] (at. %)	H_{dc} (kOe)	H_{osc} (kOe)	τ_0 (min)	p_{H}	$\bar{p}_{es}$	ϵ_{eff}
$13\times15\times3$	0.9×10^{-4}	2.85	4.60	13.3	0.032	0.348	3.83
		2.85	6.30	13.3	0.072	0.459	3.31
		2.84	6.30	11.8	0.063	0.453	3.58
		2.84	8.50	13.3	0.126	0.582	3.16
$13\times15\times9$	0.9×10^{-4}	2.87	6.30	12.5	0.065	0.459	3.47
		6.00	6.30	19.5	0.182	0.452	2.25
		2.87	8.50	14.3	0.138	0.582	2.97
		2.89	6.30	15.0	0.073	0.459	3.15
		3.48	6.30	18.3	0.102	0.458	2.75
$8\times7\times7.5$	0.9×10^{-4}	2.85	6.30	13.7	0.072	0.459	3.27
		2.95	6.30	14.0	0.065	0.459	3.29
		2.95	4.65	11.7	0.031	0.351	3.79
		5.00	6.30	18.7	0.125	0.456	2.84
		4.00	6.30	17.3	0.115	0.458	2.76
$7.1\times7.4\times7.3$	2.6×10^{-4}	3.02	6.30	4.0	0.070	0.459	3.54
		3.02	4.65	3.7	0.032	0.351	3.93
		3.02	8.50	3.4	0.134	0.582	3.37

I, the proton polarization is virtually independent of the sample shape and size; the nonresonance heating is negligible, at least for these operating conditions.

CONSTRUCTION OF THE EQUIPMENT

For the neutron test it was decided to rotate the sample in a stationary magnetic field. This solution was thought to be technically simpler, especially since the magnetic field defines the polarization axis not only of the protons, but also of the transmitted neutrons. The equipment was built utilizing and modifying available components such as an electromagnet capable of reaching almost 15 kOe with a 2-in. pole gap, motor controls for a neutron chopper, and a conventional ^{4}He cryostat. The tail of the cryostat, as modified for the experiment, is illustrated in Fig. 5. The YES crystal is located on a shaft in a cavity which is cryogenically connected to the cryostat. Temperatures lower than 4.2 K are obtained by connecting the helium bath of the cryostat to the bottom of the sample cavity with a calibrated flow-resistive coil. The cavity is kept at low pressure by an operating pumping station. The system reaches equilibrium 6 h after filling the cryostat with liquid helium. An equilibrium pressure in the cavity of 0.6 mm Hg (or a temperature of 1.2 K) is achieved with a pumping speed of 50 ft^3/min. The volume of superfluid helium in the sample cavity is measured by the flash or overpressure created when the cryostat is emptied of 4.2-K helium, so that the sample cavity is no longer thermally protected. The data indicate that during the measurements the sample is always (irrespective of its speed of rotation) well immersed in the supercooled bath.

The sample is rotated by a synchronous motor situated at room temperature to which the sample is attached by a composite tube of fiberglass and Pyrex. Even the shape of the tube received particular attention. In order to avoid heat losses due to friction, the tube is free standing. Its designed resonance frequency was 180 Hz; tests performed with a vibration pickup attached to the spring-suspended motor indicated that the rotation was extremely smooth up to 150 Hz. This was kept as the maximum frequency of operation. The motor was controlled by a phase-sensitive device, and reached and locked in the preset rotational speed after operating in the slip mode for a few seconds.

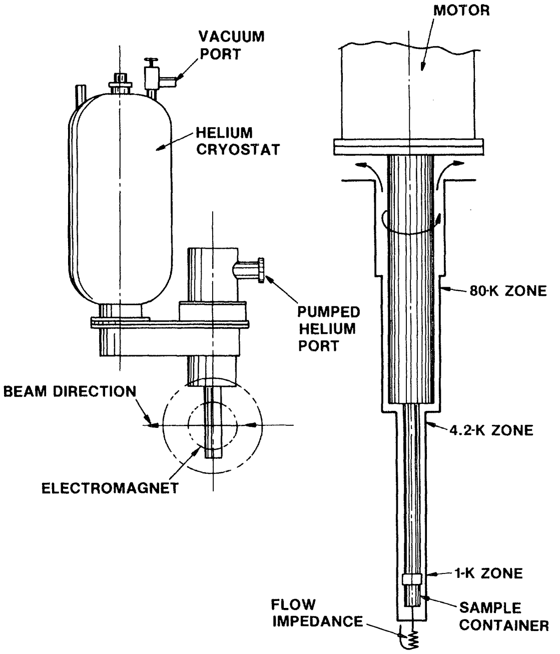

FIG. 5. Scheme of the spin-refrigerator cell used for the neutron-transmission experiment.

The YES crystal was attached to the bottom of the rotating tube in a self-pumping container of Kel-F (Minnesota Mining and Manufacturing Company). The crystal was in the shape of a cylinder (1 cm in diameter, 1 cm long) with its axis normal to the hexagonal *c* axis.

In the original design of the apparatus, a superconducting quantum-interference device (SQUID) susceptometer was included to independently measure the nuclear magnetization of hydrogen. Actually this "broad-band" probe should have measured the oscillating paramagnetic signal of Yb^{3+} as well as the slow increase of the proton magnetization (the diamagnetic response of the YES matrix was found to be quite isotropic). Unfortunately, the SQUID system proved to be too noisy when the motor was turned on, and no method was found to effectively shield the magnetic noise. Otherwise, the system functioned very well, both from the mechanical and cryogenic standpoint, operating continuously for weeks with very little maintenance, and with a helium consumption of 12 l/d.

NEUTRON TESTS

The neutron-transmission measurements were performed at the Intense Pulsed Neutron Source at Argonne National Laboratory, Argonne, Illinois. The sample was located in the neutron beam 6 m from the source, and the transmitted-beam intensity was measured by a ^{3}He detector at a distance of 1 m from the sample. The cryostat assembly was set with the magnetic field perpendicular to the neutron beam, which had to cross only the tail of the cavity, which was made entirely of neutron-transparent aluminum. The neutron beam was defined by collimators in the neutron-shielding monolith and by thick boron nitride masks (of 6×6 mm^2 aperture) before and after the sample. This was aligned by optical means and eventually by neutron radiography.

In view of the thickness of the sample (~1 cm) the neutron-transmission measurements are more sensitive to the proton polarization at the cold end of the spectrum; the unwanted radiation was removed by placing a cold beryllium filter in front of the beam port. A typical transmission spectrum is shown in Fig. 6. Since the pulsed source delivers neutron bursts ~10 μsec long (with a frequency of 30 Hz) the wavelength of the neutrons arriving at the counter is determined by time-of-flight measurements.

The measurement of the transmitted spectrum with and without the sample (rotating in zero magnetic field, or unpolarized) enabled the determination of the total cross section of the crystal. This, recast as the microscopic cross section for hydrogen, is presented in Fig. 7. The compar-

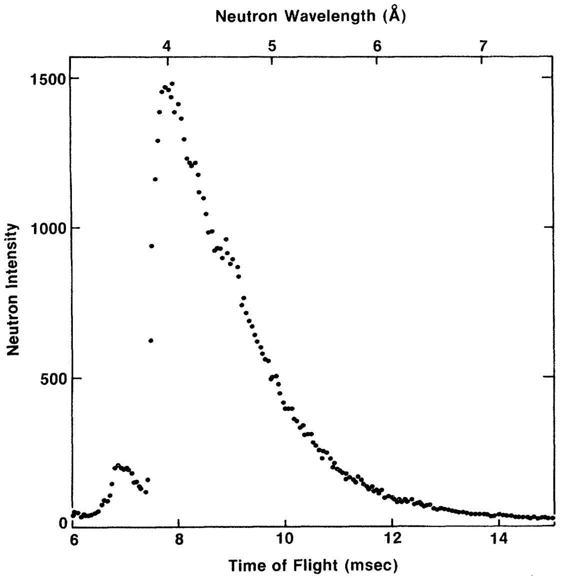

FIG. 6. Typical neutron spectrum from the pulsed neutron source at Argonne after being transmitted through a beryllium filter and 1-cm-thick YES crystal. The sharp edges are entirely due to beryllium and the aluminum structures. The neutron intensity is for a time interval of 50 μsec, in one hour of machine operation.

ison with the accepted value of the scattering cross section is almost immediate, since the other cross sections in YES are relatively small. Absorption in YES accounts for 0.89×10^{-24} cm^2/H (4-Å neutrons); the incoherent scattering from nuclei other than hydrogen is negligible, and the diffraction effects were also assumed to be negligible. The corrected data points are quite close, at the low-energy end, to the limit $\sigma_0=79.7\times10^{-24}$ cm^2 presented in Eq. (2). There is, however, still an energy dependence $\sigma(E)$, and this opens the question of whether or not it is legitimate to calculate the proton polarization from transmission data with the aid of Eqs. (2) and (3) by simply substituting σ_0 with the experimentally determined $\sigma(E)$.

An energy dependence of the hydrogen cross section is expected if the atoms are located in a potential well of finite width. The differential cross section for a given neutron wavelength can be expressed as a Debye-Waller factor.[19] Its integration over all scattering angles gives

$$\sigma(E)=\sigma_0\int_0^{\pi}\exp(-2cE\overline{u^2}\sin^2\theta)\sin2\theta\,d\theta$$
$$\simeq\sigma_0(1-cE\overline{u^2})\ . \qquad (12)$$

Equation (12) is explicitly written in terms of the neutron energy (in meV). The constant c is 8.92 Å^{-2}meV^{-1}, and $\overline{u^2}$ is the mean-square displacement of the hydrogens in the ground state. The form of Eq. (12) is in reasonable agreement with the trend of the data. However, a least-squares-fitting procedure gives $\sigma_0=82.2\times10^{-24}$ cm^2 and $\overline{u^2}=0.025$ Å^2: The extrapolated cross section is larger than expected, while the mean-square displacement is approximately twice the normal value for hydrogen in organic materials. Rigorously speaking, inelastic terms must be present, and these change the form of the polarization dependence of $\sigma^{\pm}$. In practice, these terms can be neglected. Several transmission runs with the crystal rotating in a magnetic field were taken with sufficient accuracy to obtain the proton polarization at different neutron energies. The polarization, determined with the aid of the

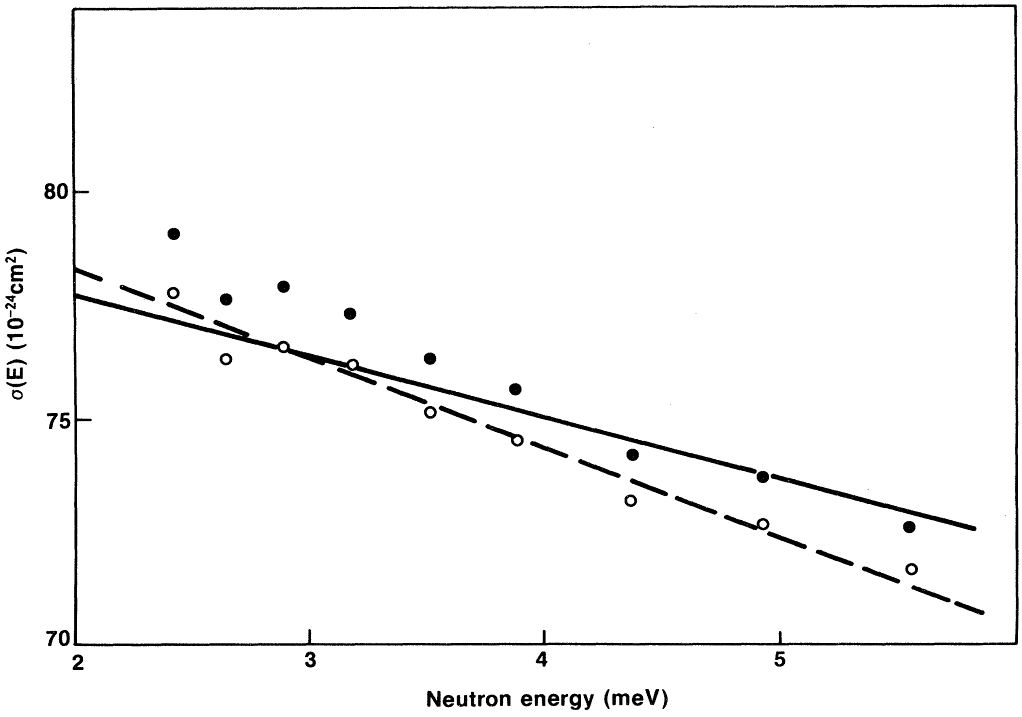

FIG. 7. Solid circles: experimental cross section for hydrogen in YES. Open circles: experimental cross section corrected for absorption. Dotted line: least-squares-fitted $\sigma(E)$ with the form given by Eq. (12). Solid line: expected behavior of $\sigma(E)$ for $\sigma_0=79.7$ b and a mean-square displacement of hydrogen $\overline{u_s^2}=0.013$ Å^2.

experimental $\sigma(E)$, was, in all cases, constant over the entire range of neutron energies used.

Table II presents a compendium of the proton polarization obtained by neutron transmission. The measurement of an experimental rise time τ_0 in this case was difficult in view of the relative slowness in collecting neutrons to obtain adequate statistics. Only for the ytterbium-poor sample was it possible to arrive at estimate of $\tau_0 \sim 30$ min, fairly close to the value of $\tau N_H/N_e$ which, at the rotating speed of 150 sec^{-1}, is 20 min. The temperature indicated in Table II is that of the helium bath; however, the temperature of the sample might have been higher due to imperfect cooling.

A fair amount of effort was spent in testing samples in conditions in which the cooling might have been different. Usually the samples were coated with Kel-F grease to protect them from dehydration during the cool down, but some runs were carried out with uncoated samples. Other crystals had small holes drilled in them to facilitate cooling, and one sample was rotated virtually without a container. In no case was the nuclear polarization improved. Finally, sample 1 (cf. Table II) was crushed into granular form. The polarization found for this randomly oriented aggregate was lower by approximately 25% than that for the single crystal, as was expected[12] (it should be noted that the transmission measurement is inherently less accurate for an aggregate than for a uniform sample since the transmission has to be averaged over quite different sample thicknesses).

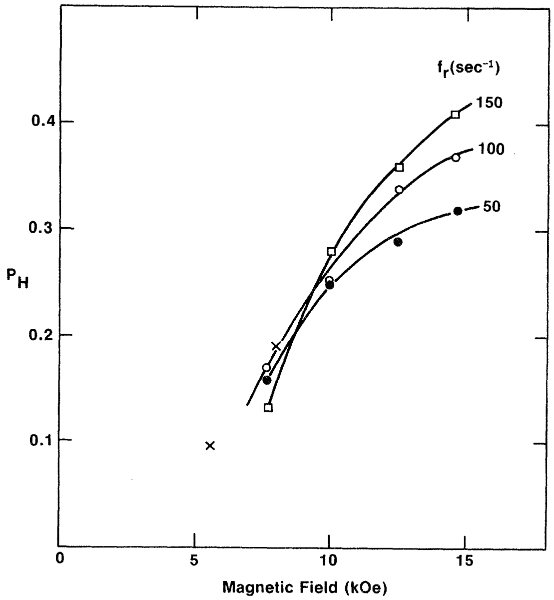

FIG. 8. Proton polarization measured by neutron transmission (sample 1, $[\text{Yb}]/[\text{Y}]=2.6\times10^{-4}$ at. % at $T=1.21$ K). The statistical accuracy per point is $\pm1\%$ proton polarization. Curves drawn to join the data points at the same rotation frequency. $\times$ denotes proton polarization measured by NMR with the sample in a rotating field at $f_r=25$ sec^{-1} and $T=1.3$ K.

The values of the proton polarization reported in Table II, to be taken with a statistical uncertainty of 1%, indicate that the response of two samples is identical. The proton polarization increases rapidly with the field and the rotation frequency; it increases also (in the narrow temperature range available) with a better cooling of the sample. The data for one of the samples are presented in graphical form in Fig. 8, together with the proton polarization obtained at lower fields by the NMR method. The two types of measurements seem to be in substantial agreement.

DISCUSSION

The best result obtained, 41% proton polarization, is not yet sufficiently high to make the spin refrigerator a viable neutron-spin filter. This is obvious by observing (in

TABLE II. Proton polarization in YES rotating in a magnetic field. Neutron-transmission measurements for 1-cm-thick crystal. Units of concentration are at. %.

	H_1 (kOe)	f_r (sec^{-1})	T (K)	Transmission ($E=2$–5 meV)	p_H	$\bar{p}_{es}$
Sample 1	0	50–150	1.22	0.0154	0	0
$[\text{Yb}]/[\text{Y}]=2.6\times10^{-4}$	7.65	50	1.22	0.0177	0.16	0.43
	10	50	1.21	0.0208	0.25	0.50
	12.5	50	1.21	0.0229	0.29	0.49
	14.7	50	1.21	0.0246	0.32	0.46
	7.65	100	1.21	0.0178	0.17	0.44
	10	100	1.21	0.0210	0.25	0.53
	12.5	100	1.21	0.0263	0.34	0.57
	14.6	100	1.21	0.0292	0.37	0.55
	7.75	150	1.21	0.0168	0.13	0.44
	10	150	1.21	0.0223	0.28	0.54
	12.5	150	1.21	0.0279	0.36	0.60
	14.6	150	1.21	0.0327	0.41	0.61
	14.6	150	1.29	0.0281	0.36	0.57
Sample 2	0	150	1.21	0.0157	0	0
$[\text{Yb}]/[\text{Y}]=0.9\times10^{-4}$	12.9	150	1.21	0.0284	0.36	0.60
	13.2	150	1.14	0.0332	0.41	0.64

Table II) that the neutron transmission is still rather low; the polarization of the transmitted cold neutrons is estimated to be 84%. It is also evident from Fig. 8 that significantly higher proton polarization can be achieved by rotating the sample faster in a higher magnetic field; some estimate can be made about the new requirements. At $H=14.6$ kOe the term K_e appearing in the expression for the proton polarization, Eq. (9), is unitary, and thus we have simply $p_H=\bar{p}_{es}/\epsilon_{\text{eff}}$. If the temperature of the sample is that of the helium bath, then the electronic polarization takes the value presented in Table II. From these values, it appears that at $H=14.6$ kOe, $\epsilon_{\text{eff}}=1.48$, irrespective of the rotation frequencies; the coupling is much better than that measured by NMR at lower H and is still improving with fields. However, p_{H} should show a large increase with increasing rotational frequency if the behavior of the multiple spin-flip probabilities is as predicted.[12] The acceptance of this argument might indicate that the temperature of the sample in our neutron experiment is higher than 1.2 K with ΔT proportional to the rotational frequency. According to this reasoning, at higher f_r the electronic polarization $\bar{p}_{es}$ and $\epsilon_{\text{eff}}=\bar{p}_{es}/p_{\text{H}}$ are lower. If, for example, the effective temperature is $T=1.5$ K, $\epsilon_{\text{eff}}=1.21$ for $f_r=150$ sec^{-1} and $H=14.6$ kOe. This temperature is also the maximum temperature that can be assigned to the sample; otherwise $p_{\text{H}}(H)$ would exhibit an approach to saturation that is not experimentally observed. If the preceding analysis is correct, considerably higher polarization will be obtained for the same values of H and rotational frequency for a true sample temperature of 1.2 K.

With all the uncertainties of the present analysis, we conclude that a proton polarization of 60–65 % should be attainable with rotational frequencies of less than 300 sec^{-1}, a magnetic field less than 25 kOe, and somewhat improved sample cooling. These conditions (which are also approximately the limits of the currently acquired technology) should enable the operation of an adequate neutron-spin filter.

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[1] G. E. Bacon, *Neutron Diffraction,* 3rd ed. (Clarendon, Oxford, 1975).

[2] V. I. Lushchikov, Yu. V. Taran, and F. L. Shapiro, Yad. Fiz. 10, 1178 (1969) [Sov. J. Nucl. Phys. 10, 669 (1970)].

[3] F. F. Freeman and W. G. Williams, J. Phys. E 11, 459 (1978).

[4] A. Abragam and M. Goldman, Rep. Prog. Phy. 41, 395 (1978), and references therein.

[5] *Proceedings of the International Conference on Polarized Targets and Ion Sources, Saclay, France, 1966* (Le Documentation François, Paris, 1967); *High Energy Physics with Polarized Beams and Polarized Targets (Argonne, 1978),* Proceedings of a Symposium, edited by G. H. Thomas (AIP, New York, 1979).

[6] G. A. Keyworth, J. R. Lemley, C. E. Olsen, F. T. Seibel, J. W. T. Dabbs, and N. W. Hill, Phys. Rev. C 8, 2352 (1973).

[7] G. T. Jenkin, Institut Laue Langevin Technical Report No. 77 JE44T (unpublished).

[8] S. Hiramatsu, S. Isagawa, S. Ishimoto, A. Masaike, K. Morimoto, S. Funahashi, Y. Hamaguchi, N. Minakawa, and Y. Yamaguchi, J. Phys. Soc. Jpn. 45, 949 (1978).

[9] A. Abragam, Cryogenics 3, 42 (1963).

[10] C. J. Jeffries, Cryogenics 3, 41 (1963).

[11] K. H. Langley and C. D. Jeffries, Phys. Rev. 152, 358 (1966).

[12] J. R. McColl and C. D. Jeffries, Phys. Rev. B 1, 2917 (1970).

[13] Wendell H. Potter and H. R. Stapleton, Phys. Rev. B 5, 1729 (1972).

[14] H. B. Brom and W. J. Huiskamp, Physica 66, 43 (1973).

[15] J. Button-Shafer, R. L. Lichti, and W. H. Potter, Phys. Rev. Lett. 39, 677 (1977).

[16] J. P. Wolfe and C. D. Jeffries, Phys. Rev. B 4, 731 (1971).

[17] R. W. Broach, J. M. Williams, G. P. Felcher, and D. G. Hinks, Acta Crystallogr. Sect. B 35, 2317(1979).

[18] J. A. A. Ketelaar, Physica 4, 619 (1937).

[19] R. W. James, *The Optical Principles of the Diffraction of X-Rays* (Cornell University Press, Ithaca, New York, 1965).