Depolarization of neutrons in ferromagnetic holmium by means of enhanced nuclear parity violation in ¹³⁹La

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The depolarization of epithermal neutrons in a thick single crystal of ferromagnetic holmium has been measured and analyzed with a model of neutron precession through a highly correlated array of magnetic domains. The large parity violation in the 0.734-eV *p*-wave resonance in ¹³⁹La was used to analyze the neutron polarization, and represents an application of parity violation in nuclear resonances to a measurement in condensed-matter physics.

The large fluxes of epithermal neutrons available from spallation sources make possible new types of neutron scattering measurements in nuclear and solid-state physics. However, the use of polarized epithermal neutrons is limited because experiments require a filter material itself polarized either by cryogenic or optical means. In this paper a monoenergetic analyzer based on parity violation in the neutron-nucleus reaction will be described which does not require a complicated polarized filter. Normally parity violation in the nucleus is very small, but in certain nuclear resonances the effect is enhanced to as large as several percent. In the present experiment this enhanced parity violation has been used as an analyzer for a measurement in condensed-matter physics—the depolarization of neutrons in a thick ferromagnetic crystal.

We have measured the neutron depolarization through the 2.29-cm-thick single crystal of ferromagnetic holmium target using a polarized epithermal neutron beam at the Los Alamos Neutron Scattering Center (LANSCE). The enhanced parity violation in the 0.734-eV resonance in ¹³⁹La was used to analyze the polarization of the neutron beam. We outline below how this unusual nuclear phenomenon was applied to a condensed-matter measurement. We first describe the internal magnetic fields in holmium, models of neutron depolarization in solids, and then the experimental measurement.

The magnetic structure of holmium has been extensively studied by magnetization experiments¹ and neutron scattering.^{2,3} Below 20 K the hexagonal crystal forms a helical ferromagnetic phase in which the *c* crystalline axis is the screw axis and the electronic moments have a turn angle of 30° per layer. The moments are canted 10° out of the *a-b* plane, producing a net magnetic moment of 1.7 Bohr magnetons parallel to the *c* axis. A large magnetic field must be applied in the *c* direction to rotate the moments parallel to the *c* axis, but a much smaller field applied in the *a-b* plane collapses the spins into this plane. The saturation internal field in the *c* direction is about 0.6635 T and in the *a-b* plane 3.87 T.

The domain structure of holmium is less well described. There have been no studies of thick single crystal samples. Domains in thin single crystals of holmium have been imaged by a polarized neutron diffraction tomography technique.⁴ The average size of the domains was very sensitive to crystal purity, as parametrized by the residual resistivity ratio R(300 K)/R(4 K). The average domain size decreased below the resolution of the tomography technique (60 μ m) when the resistivity ratio was less than 50. The observed domain walls had no simple crystallographic orientations and the shapes of the domains were insensitive to the thermal history of the crystal. This agrees with the observation that the domain structure should be stable because the chiral nature of the magnetic ordering dominates over the ferromagnetic aspect.5

The depolarization of polarized neutrons passing through a ferromagnetic target has been discussed by Halpern and Holstein⁶ and applied to polycrystalline holmium by Postma *et al.*⁷ Classically a neutron passes through a set of ferromagnetic domains and precesses in the internal magnetic field of each domain. The precession of a neutron in a single magnetic domain of thickness δ and having an internal field component B_1 perpendicular to the neutron spin is

$$\Delta\theta = \gamma_n B_\perp \delta / v_n ,$$

where γ_n is the neutron gyromagnetic ratio $2|\mu_n|/\hbar$ and v_n is the neutron velocity.

In the quantum picture the polarization of the incident neutron beam is

$$f_{n0} = \frac{N^+ - N^-}{N^+ + N^-} ,$$

where N^+ and N^- are the numbers of neutrons having spin parallel or antiparallel to the quantization axis. The polarization of the neutron beam exiting the target is similarly denoted f_n . The depolarization of the beam in a polycrystalline flat slab of thickness t is described as $f_n/f_{n0} = \exp(-2D't)$ where D' is the depolarization parameter, the inverse of the mean free path for a spin reversal. In the limit where the precession in each domain is relatively small and the domains are magnetized in random directions⁶

$$D'_{\rm random} = \frac{\gamma_n^2}{4} \langle B_\perp^2 \delta^2 \rangle_{\rm avg} \frac{n}{v_n^2} , \qquad (1)$$

where n is the number of domains per unit length. Note that D'_{random} is inversely proportional to the neutron kinetic energy. Because of this thermal neutron depolarization measurements are performed typically with thin samples.

In the present case, a thick single crystal, the domains in the bulk of the crystal are not magnetized in random directions. Instead they have saturated magnetizations parallel or antiparallel to the c crystalline axis. Since the net magnetization of the crystal is small there are approximately equal volumes of c-axis mirror domains. In our experiment longitudinally polarized neutrons passed through a succession of domains polarized up or down as shown in Fig. 1. The polarization of the exiting neutron beam was measured parallel to the axis of the beam. The total flux of neutrons is composed of many thin beams which traverse different sets of domains in the crystal. After traversing a set of N domains the polarization of such a thin beam is

$$f_n = \mathbf{D}(\mathbf{n}_N, t_N) \mathbf{D}(\mathbf{n}_i, t_i) \cdots \mathbf{D}(\mathbf{n}_1, t_1) f_{n0}$$
,

where each $\mathbf{D}(\mathbf{n}_i, t_i)$ is a 3×3 rotation matrix describing the change in the polarization occurring as the neutrons traverse one domain.^{8,9} The vector \mathbf{n}_i describes the orientation of the local field in the *i*th domain relative to the neutron polarization direction and t_i is the time the neutron spends in the domain. For a set of domains having internal magnetic fields B_i canted at an angle θ to the neutron beam direction the component of the neutron polarization along the beam is given by

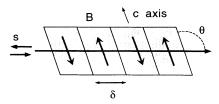


FIG. 1. Schematic of the depolarization of a longitudinally polarized neutron beam (s), of momentum k incident on a set of magnetic domains polarized parallel or antiparallel to the c crystalline axis. In the model calculation in the text it is assumed that the domain thickness δ is independent of θ .

$$f_n / f_{n0} = \cos^2 \theta + \sin^2 \theta \langle \cos \Phi \rangle$$
,

where

$$\Phi = \sum_{i}^{N} (-1)^{i} \left[\frac{\gamma_{n} B_{i} \delta_{i}}{v_{n}} \right] .$$

The $(-1)^i$ term indicates that the sense of the neutron precession is opposite for domains polarized in the "up" or "down" directions. The $\cos^2\theta$ term indicates that the component of the neutron polarization parallel to the domain magnetization axis is preserved in transmission through the crystal. The quantity $\cos\Phi$ is averaged over all paths of neutrons through the crystal. The net depolarization of the beam therefore depends upon the $\langle \cos\Phi \rangle$ term, i.e., different parts of the total neutron flux traverse different sets of "up" and "down" domains.

A simple model (Fig. 1) illustrates the relation of $\langle \cos \Phi \rangle$ to the distribution of domain magnetizations. For domains of uniform thickness δ , the neutron beam traverses $N=t/\delta$ domains in a target of thickness t. Over the entire crystal the average number n^+ of "up" domains is equal to the average number n^- of "down" domains but for any particular neutron path the quantity $\Delta n = n^+ - n^-$ is a random variable centered about zero. Assuming a binomial distribution of the numbers of up and down domains, with N large, and assuming the precession in each domain is small, then

$$\langle \cos \Phi \rangle = \int_{\Delta n = -N}^{\Delta n = N} \cos \left[\frac{\gamma_N B_i \delta}{v_N} \Delta n \right] P(\Delta n) d(\Delta n) ,$$

and $P(\Delta n)$ is given by

$$P(\Delta n) = \left(\frac{2}{N\pi}\right)^{1/2} \exp\left\{-\frac{2(\Delta n)^2}{N}\right\}.$$

In the limit $N \rightarrow \infty$ the integral becomes

$$\langle \cos \Phi \rangle = \exp \left\{ - \left[\frac{\gamma_N B_i}{2\sqrt{2}v_N} \right]^2 \delta t \right\}$$

 $\equiv \exp\{-2D'_{\text{bulk}}t\}$.

The depolarization of the neutron beam is such a sample having internal fields aligned along one direction is therefore

$$f_n / f_{n0} = \cos^2 \theta + \sin^2 \theta \{ \exp(-2D'_{\text{bulk}}t) \} .$$
⁽²⁾

The exponential term is similar to the depolarization of neutrons in a polycrystalline target, but derives here from the randomness in the number of c-axis domains traversed. In the polycrystalline target the domains are also magnetized in random directions.

Basic to any depolarization measurement is the determination of the polarization of the neutron beam. In the present work we have used a technique applicable at epithermal neutron energies which does not require polarized targets or cryogenics. In certain compound nuclear resonances parity-violating (PV) effects are significantly enhanced, as demonstrated in the 0.734-eV *p*-wave resonance in ¹³⁹La. The cross section for neutrons polarized parallel (+) or antiparallel (-) to the beam momentum

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is $\sigma_{\pm} = \sigma_0(1 \pm f_n P)$ where σ_0 is the helicity independent resonance cross section. At 0.734 eV the value of the parity violation P, 0.1015±0.0045, is large enough to provide an accurate method of determining the longitudinal polarization of the neutron beam. Other such parityviolating resonances which may be exploited for polarization analysis are given in Table I. An approximate figure of merit for polarization analysis is the product $\sigma_0 P$. Known resonances that show parity violation occur at many different energies, from 0.7 to about 80 eV. Although not as efficient as the Bragg reflection polarizersanalyzers used at lower neutron energies,⁹ the parityviolation analyzer should be useful in other solid-state measurements which require polarized epithermal neutrons.

The holmium target was a cylindrical crystal, 99.8% purity, 2.29 cm in diameter and 2.8 cm long, 0.62 moles of holmium grown and shaped at the Materials Preparation Center at the Ames Laboratory of Iowa State University. The crystal was selected because it had been used for investigations of fundamental symmetries in neutron transmission.¹⁰ The sensitivity of such experiments can be degraded by precession of the neutron spin in the interior of the crystal. The c crystallographic axis as determined by x-ray diffraction was oriented perpendicular to the cylindrical axis. This orientation was verified to $\pm 1^{\circ}$ in nuclear deformation experiments at the Triangle Universities nuclear laboratory (TUNL).¹¹ The resistivity of the target was measured parallel to the cylinder axis (in the a-b plane). The residual resistivity ratio was $R_{293 \text{ K}}/R_{4 \text{ K}} = 40$, therefore the average domain size is expected to be less than 60 μ m.

The experimental setup and data analysis were similar to that used for recent measurements of parity violation in ¹³⁹La.¹² As shown in Fig. 2 a beam of epithermal neutrons from the LANSCE source was longitudinally polarized by filtering through a cryogenic polarized proton

TABLE I. Compound nuclear resonances displaying large enhancements of parity violation.

Isotope	<i>E</i> (ev)	$P(10^{-3})$	$\sigma_0(b)^a$	$\sigma_0 P (10^{-3})$
¹³⁹ La	0.73	100±4 ^b	2.8	280
⁸¹ Br	0.88	$24\pm4^{\circ}$	0.9	22
¹¹⁷ Sn	1.33	4.5±1.3°	1.6	7.2
¹¹¹ Cd	4.53	$-8.2\pm2.2^{\circ}$	3.8	31
²³⁸ U	63.5	25 ± 4^{d}	10.1	250
²³² Th	128.2	13.1 ± 1.8^{e}	56	730

^aCalculated from parameters in S. F. Mughaghab, M. Divadeenam, and N. E. Holden, *Neutron Cross Sections* (Academic, New York, 1981).

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Beam Dynamically Spin Neutron Polarized Flipper Counters Spallation Proton Target Source Cryostat

Holmium

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in Ćryostat

Target

FIG. 2. Schematic of beam and target geometry showing locations of holmium target and lanthanum analyzing filter.

target. The beam passes through the holmium target, an adiabatic spin flipper, and a ¹³⁹La target before reaching a detector located 25 m from the neutron source. The neutron spins are flipped every 10 sec. The polarization of the 0.734-eV neutrons was determined by measuring the parity-violating asymmetry

$$\varepsilon = \frac{Y_+ - Y_-}{Y_+ + Y_-} = -\tanh(f_n n t \sigma_0 P) ,$$

where Y_{+-} is the neutron transmission yield at the detector for the two spin orientations, *n* is the number density of the ¹³⁹La target and *t* the ¹³⁹La target thickness.¹² This gives a direct measure of the polarization of the neutron beam after exiting the holmium sample. About two hours of running time were required to measure f_n/f_{n0} for each *c*-axis orientation θ with respect to the beam direction.

The holmium target was affixed to the end of a copper coldfinger attached to a liquid-helium reservoir. The crystal was held in vacuum and the neutron beam passed through thin aluminum walls of the Dewar at room temperature and 77 K. The ambient magnetic field due to the polarizer and the spin flipper at the location of the crystal was 100 G. Because of the large shape demagnetization factor of the cylinder we assume that the net crystal magnetization was vanishingly small.

The target was cooled to 4 K and inserted into the beam line with the c axis oriented perpendicular to the direction of the neutron momentum and polarization. The cryostat and target were then rotated in place to the next angle θ in the order listed in Table II. The target was then warmed into the paramagnetic phase at room temperature and the depolarization was measured with the c axis perpendicular and parallel to the neutron beam. The warm target data show no depolarization due to the paramagnetic phase of the holmium. To check for magnetic hysteresis the target was again cooled to 4 K in the ambient field in another room and placed in the beam line with the c axis parallel to the beam direction. As the target was slowly moved into position the cryostat was rotated so that the c axis was always parallel to the local magnetic field.

The overall dependence of the depolarization on the orientation of the crystal indicates that the bulk domains are very well correlated along the c axis. With the neu-

Lanthanum

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Neutron

Target

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Angle (deg)	f_n/f_{n0}	Temperature (K)
No target	1.0 (Calibration)	
90	-0.076 ± 0.142	4
0	$+0.685 \pm 0.062$	4
45	$+0.432\pm0.076$	4
30	$+0.699\pm0.180$	4
60	$+0.249\pm0.076$	4
15	$+0.65\pm0.075$	4
0	$+0.97\pm0.051$	300
90	+1.01	300
0	+0.64±0.12	4

TABLE II. Summary of depolarization data.

tron spins parallel to the c axis $(\theta=0^{\circ})$ the beam is transmitted with about 30% depolarization. On the other hand, the beam is essentially completely depolarized in the transverse geometry $(\theta=90^{\circ})$. However, since $f_n/f_{n0}\neq 1$ at $\theta=0^{\circ}$ there must be depolarization besides that described by Eq. (2).

The data can be analyzed as the product of depolarization by the correlated bulk *c*-axis domains and another depolarization caused either by closure domains¹³ or by local variations in the *c*-axis direction. Variations in the *c* axis can be modeled as random fields as in Eq. (1), so that

$$\frac{f_n}{f_{n0}} = \mathbf{D}_{\text{bulk}} \mathbf{D}_{\text{random}}$$
$$= [\cos^2\theta + \sin^2\theta \exp(-2D'_{\text{bulk}}t)] \exp(-2D'_{\text{random}}t) .$$
(3)

A fit of this form is shown as the solid line in Fig. 3 using $D'_{\rm bulk} = 0.53$ /cm and $D'_{\rm random} = 0.11$ /cm. The quantity t was taken to be the average chord length of the cylindrical target, 1.806 cm.

A slightly better fit to the data is obtained if it is assumed that there is depolarization from closure domains, which would have an internal magnetic field in the *a-b* plane perpendicular to the magnetization of the bulk domains. The depolarization can be calculated following the derivation of Eq. (2) giving

$$\frac{f_n}{f_{n0}} = \mathbf{D}_{\text{bulk}} \mathbf{D}_{\text{closure}}$$
$$= [\cos^2\theta + \sin^2\theta \exp(-2D'_{\text{bulk}}t)]$$
$$\times [\sin^2\theta + \cos^2\theta \exp(-2\Delta)], \qquad (4)$$

where $\Delta = D'_{a-b} t_{\text{closure}}$. D' is the depolarization parameter for the closure domains, and the net thickness t_{closure} of these domains is unknown. This form is also displayed as the dotted line in Fig. 3, using $\exp(-2D'_{\text{bulk}}t)=0$ and

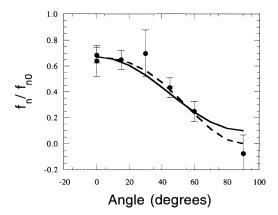


FIG. 3. Neutron depolarization as a function of the angle θ between the *c* axis and neutron polarization direction. The solid line is a fit to Eq. (3) with depolarization due to both bulk magnetic domains and variations in the *c*-axis direction. The dotted line is a fit to Eq. (4) with depolarization due to both bulk magnetic domains and closure domains.

 $\Delta = 0.20$. The precision of the data is not sufficient to distinguish whether closure domains, Eq. (4), or *c*-axis variations, Eq. (3), cause the additional depolarization.

Although the average size of the bulk domains cannot be determined from these measurements, a lower limit of $\delta \ge 11 \ \mu m$ is given from $f_n / f_{n0} \ (\theta = 90^\circ) \le 0.14$ and Eq. (3). The quantity δ could be determined by similar measurements at higher neutron energies, using other resonances as analyzers.

We have measured the depolarization of 0.734-eV polarized neutrons in a single crystal of holmium. The enhanced parity-violation effect in ¹³⁹La was used as a polarization analyzer. The depolarization in the crystal was dependent on the relative orientation of the neutron polarization and the *c* crystalline axis. The effect was modeled in terms of neutron precession in an array of highly correlated domains magnetized in the *c*-axis direction. The results show the high degree of correlation of magnetic domains in the single crystal and demonstrate the effectiveness of epithermal neutrons in measuring properties of thick crystals. The use of other parityviolating resonances should allow polarization analysis of neutrons over a range of energies.

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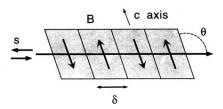


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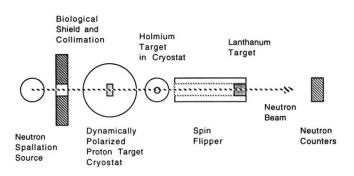


FIG. 2. Schematic of beam and target geometry showing locations of holmium target and lanthanum analyzing filter.