Interaction of Polarized Neutrons with Polarized Mn^{55} Nuclei

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(Received November 16, 1953; revised manuscript received February 15, 1954)

The dependence of the capture cross section of polarized Mn^{55} nuclei for polarized neutrons upon relative orientations of incident and bombarded particles has been directly observed. The target material was the paramagnetic substance, manganous ammonium sulfate, which is known to have a large hyperfine structure coupling. It was placed in a magnetic field of 2350 oersteds at a temperature of 0.20°K. Under these conditions the polarization of the paramagnetic electrons is about 85 percent. Because of the large effective magnetic field created by the paramagnetic electrons at the Mn nucleus, the nuclei should achieve a polarization of 16 percent. The 2.6-hour activity of the residual nucleus, Mn^{56}, was measured after the sample had been bombarded with a beam of slow neutrons polarized to the extent of 32 percent by passage through magnetized iron. The activity for neutron polarizing field and sample polarizing field parallel was found to be 3.4 percent less than for the fields antiparallel. The difference in the two activities was found to depend upon the sample temperature in accordance with theory. The difference was found to be unaccompanied by a corresponding change in sample transmission. These results are interpreted to mean that the change in sample activity was due to the dependence of the capture cross section of the polarized Mn nuclei upon the relative orientation of the interacting particles. The observations are discussed in terms of available information about the energy level system of the compound nucleus, Mn^{56}.

INTRODUCTION

THE spin dependence of nuclear forces was first demonstrated experimentally by the scattering of neutrons from ortho- and para-hydrogen,[1–4] in which experiment the effects observed are caused by the relative spin directions taken by the hydrogen atoms of each isolated hydrogen molecule. The spin dependence of scattering is exhibited also in the spin incoherent scattering of neutron diffraction experiments,[5] in which the random orientations of the nuclear spins of the sample cause an isotropic diffuse background in diffraction patterns. In both of these methods it is the spin dependence of the scattering cross section which is observed, and the bulk nuclear magnetization of the sample is zero. We have observed directly the spin dependence of the *capture* cross section by bombarding a sample possessing a bulk nuclear magnetization with a beam of polarized neutrons, and observing the dependence of the target activity upon relative spin orientations of the incident and bombarded particles. (The angular distribution of gamma rays from such polarized samples containing radioactive nuclei has been observed.[6] An experiment in which neutron effects of nuclear polarization in gadolinium sulfate were observed, has been mentioned briefly,[7] but no details of the experiment have been reported.)

For polarizing the nuclei we relied upon the method of Rose[8] and Gorter,[9] i.e., the use of the strong magnetic field produced at the nucleus by hyperfine structure coupling in an appropriate paramagnetic substance. An external magnetic field is applied to the sample merely for the purpose of polarizing the electronic moments. The sample used in this work was the deuterated paramagnetic salt, $Mn^{55}SO_4\cdot(ND_4)_2SO_4\cdot 6D_2O$. Its Mn^{55} nuclei were polarized by partial adiabatic demagnetization of the salt. This substance was selected because its hyperfine structure is known to be large,[10] its low-temperature properties are suitable, the 13-barn neutron capture cross section of Mn^{55} is acceptably large, and the 2.60-hour half-life of the radioactive compound nucleus, Mn^{56}, is convenient.

The electronic system of the Mn^{++} ion possesses spin angular momentum only. The nuclear polarization is given by

$$f_N=\frac{S(I+1)hc}{3}\frac{A}{kT}\frac{M_e}{M_\infty} \tag{1}$$

in which S is the electron spin quantum number, I is the nuclear spin quantum number, k is the Boltzmann constant, T is the temperature, M_e is the magnetic moment of the electron spin system, M_∞ is the electronic magnetic moment at saturation, h is Planck's constant, c is the velocity of light, and A is the hyperfine structure coupling constant. The spatial average value of A for this substance has been calculated from measurements[10] to be 0.00932 cm^{-1}. Substituting the appropriate values $S=5/2$, $I=5/2$, expression (1) becomes

$$f_N=\frac{0.0391}{T}\frac{M_e}{M_\infty}. \tag{2}$$

[1] Halpern, Estermann, Simpson, and Stern, Phys. Rev. **52**, 142 (1937).
[2] Brickwedde, Dunning, Hoge, and Manley, Phys. Rev. **54**, 266 (1938).
[3] L. W. Alvarez and K. S. Pitzer, Phys. Rev. **58**, 1003 (1940).
[4] Sutton, Hall, Anderson, Bridge, DeWire, Lavatelli, Long, Snyder, and Williams, Phys. Rev. **72**, 1147 (1947).
[5] Shull, Wollan, Morton, and Davidson, Phys. Rev. **73**, 262 (1948).
[6] Ambler, Grace, Halban, Kurti, Durand, Johnson, and Lemmer, Phil. Mag. **349**, 216 (1953).
[7] J. A. Beun, Ned. Tijdschr. Natuurk. **18**, 245 (1952).
[8] M. E. Rose, Phys. Rev. **75**, 213 (1949).
[9] C. J. Gorter, Physica **14**, 504 (1948).
[10] B. Bleaney and D. J. E. Ingram, Proc. Roy. Soc. (London) **A205**, 336 (1951).

In this experiment the manganous ammonium sulfate sample was demagnetized from an initial temperature of 1.2°K and an initial magnetic field of 16 400 oersteds to a final magnetic field of 2350 oersteds. This demagnetization procedure gave a final temperature of 0.20°K in the final field of 2350 oersteds. The electron polarization, $f_e=M_e/M_\infty$, under these conditions of final field and temperature was measured to be $f_e=85$ percent. The cryogenic aspects of this work, including the thermodynamic measurement of the final temperature and the measurement of the sample magnetic moment will be described in detail in a separate publication.[11] Substituting the measured values $f_e=0.85$, $T=0.20$°K in (2), a nuclear polarization $f_N=16.6$ percent is to be expected.

The nuclear cross section, σ, for slow neutrons is given by[8]

$$\sigma=\sigma_1\left(1+\frac{I}{I+1}f_nf_N\right), \quad J=I+\tfrac{1}{2}, \tag{3a}$$

$$\sigma=\sigma_2(1-f_nf_N), \quad J=I-\tfrac{1}{2}, \tag{3b}$$

in which f_n is the neutron polarization, J is the angular momentum quantum number of the compound nucleus. σ_1 and σ_2 are the respective normal cross sections for f_n or f_N or both equal to zero. The polarization f_n of the incident neutron beam was measured[12] to be 32 percent. Using $f_N=0.16$, $f_n=0.32$ in (3a) and (3b), the observed Mn capture cross section for spins of neutrons and nuclei parallel should be 7.2 percent greater than the cross section for antiparallel spins, if only a level of $J=I+\frac{1}{2}$ is effective, and should be 10.2 percent less, if only $J=I-\frac{1}{2}$ is effective. If only a single resonance level is effective, and if the product of the two polarizations (f_nf_N) is known, then we can directly deduce whether the angular momentum of the compound state is $I+\frac{1}{2}$ or $I-\frac{1}{2}$ from the observation of whether the neutron cross section increases or decreases with polarization. If more than one resonance level is effective, then terms of both types (3a) and (3b) may contribute to the cross section, σ_0. The measured relative change of cross section with polarization will then have to be interpreted in terms of the nuclear parameters of the effective resonance levels.

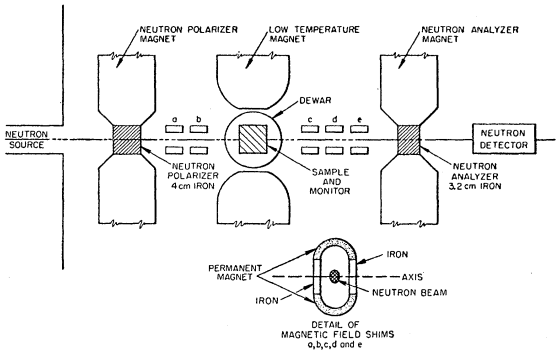

FIG. 1. Schematic arrangement of apparatus.

Our measurements of the capture cross section of Mn^{55} showed a decrease of 3.4±0.3 percent, when the relative polarization directions (spin directions) of neutrons and nuclei were changed from the antiparallel to the parallel state. The interpretation of the measured negative 3.4 percent as compared to the calculated positive 7.2 percent for $J=I+\frac{1}{2}$, and the negative 10.2 percent for $J=I-\frac{1}{2}$, in terms of present knowledge of the energy level system of Mn^{56}, is given below.

A diagram of the experimental arrangement is shown in Fig. 1. The beam of neutrons emerging from the chain reactor is polarized in passing through 4 cm of highly magnetized iron. The polarized neutrons then pass through a Mn metal monitor and the deuterated manganous ammonium sulfate sample, which is at a temperature of 0.20°K in a magnetic field of 2350 oersteds. About 60 percent of the neutrons are scattered in the sample. A small fraction of the neutrons are captured by the manganese nuclei of the sample. The sample is first exposed to the beam of polarized neutrons for 50 minutes under such magnetic field conditions that the polarizations of Mn^{55} nuclei and the neutrons are parallel. The sample is then removed from the system of Dewar flasks, and its activity is carefully measured. The procedure is later repeated with the neutron and nuclear polarizations antiparallel. The activities of the sample in the two exposures is a measure of the capture cross section of Mn^{55} in the respective arrangements of relative spin orientations of neutron and nucleus.

THE CRYOSTAT AND SAMPLE

The cryostat, which provided a simple arrangement for rapidly installing and removing the sample, was constructed entirely of metal. The sample and monitor assembly is shown in Fig. 2. The hydrogen of the sample material was replaced with deuterium in order to eliminate the depolarization of the neutron beam by the large incoherent scattering of the protons, and to make the total neutron scattering cross section of the sample as small as possible. The sample of $Mn^{++}(ND_4)_2(SO_4)_2 \cdot 6D_2O$ was prepared by recrystallizing the hydrated salt four times from heavy water. Mass spectrographic analysis showed the final sample material to be 99 percent deuterated. To preserve the deuteration from exchange with atmospheric light water, it was necessary to enclose the salt in an airtight container. The container had to meet several important requirements; namely, it must not be of metal, and it could not be made of a material which would appreciably depolarize the neutrons or be activated by them. The fluorocarbon plastic, Teflon, was used. In order to admit helium

[11] J. W. T. Dabbs and L. D. Roberts, Phys. Rev. **93**, 935 (1954).

[12] Stanford, Stephenson, Cochran, and Bernstein, Phys. Rev. **94**, 374 (1954).

exchange gas to the sample during the demagnetization procedure and have the container sealed when the sample was not in the cryostat, the container was fitted with a capillary vent which could be closed. The sample was pressed from small crystals (~0.5 mm in diameter) to within a few percent of crystal density into a right circular cylinder of 0.660-in. diameter and 0.247-in. height. The sample pellet weighed 2.96 g. A cylindrical recess 0.660-in. diameter when cold and 0.010-in. deep on the back of the Teflon box served to locate the manganese metal neutron beam monitor which was 0.660-in. in diameter and 0.060-in. thick. The common cylindrical axes of the sample and monitor coincided with the axis of the neutron beam. As is usual in demagnetization experiments, the sample was suspended on strings. In this experiment, since the sample and monitor had to be removed from the apparatus daily for counting the induced activities, the sample and the monitor were clamped into a holder which was in turn suspended by a number of cotton threads from a metal framework. The strings were arranged to give a rigid reproducible support to minimize vibration-induced heat leak to the sample and to give accurate day to day repositioning.

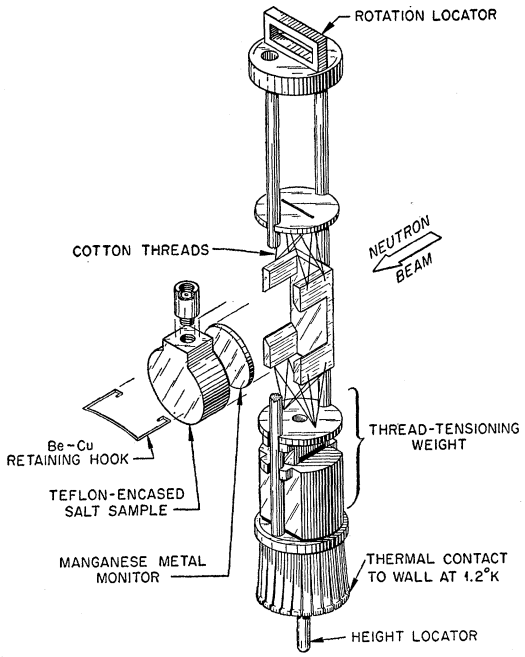

FIG. 2. Sample and monitor assembly.

POLARIZED NEUTRONS

The beam of neutrons from the reactor was polarized by passage through 4 cm of highly magnetized cold-rolled steel. The transmission method was chosen rather than polarization by total reflection from a magnetized mirror[13] or by Bragg reflection from a magnetized crystal,[14] both of which methods give a higher polarization, for reasons of intensity. The neutron polarization was reversed with respect to the polarization of the nuclei simply by reversing the neutron polarizer magnetic field with respect to the sample polarizing magnetic field. Those neutrons which did not have any collision in the sample continued on their path through the analyzer, which was 3.2 cm of highly magnetized cold-rolled steel, and then into a BF_3 proportional counter neutron detector. The direction and magnitude of the magnetic field along the path of the neutron beam were controlled by means of the parallel plate shims shown in Fig. 1, so that the neutrons should maintain the desired state of polarization. The magnetic field between the plates was adjusted by numerous U-shaped permanent magnets placed at various points along the length of the shims, as shown in Fig. 1.

For those irradiations in which the magnetic fields of neutron polarizer and sample polarizer were parallel, the fields between pairs of shims a and b were adjusted so that the magnetic field in the region between neutron polarizer and Dewar flask was at least 100 oersteds in the same direction as the two polarizing fields. Under these conditions the magnitude and direction of the neutron polarization should be maintained along the neutron path.

When the magnetic fields of neutron polarizer and sample polarizer were antiparallel, the field of shim a was adjusted to be in the same direction as that of the neutron polarizer. The field of shim b was made to be parallel to the field of the sample polarizer. The permanent U-shaped magnets were adjusted so that the magnetic field reversed its direction in the small space between the adjacent ends of shims a and b. These conditions should induce the neutrons to make non-adiabatic transitions and strike the polarized nuclei of the sample with the opposite relative orientation to that described above when the field directions of neutron polarizer and sample polarizer were parallel. In order to obtain optimum conditions for nonadiabatic transitions or no transition depending upon the type of irradiation desired, magnetic field traverses were made with flip coil and compass in adjusting the strength and direction of the magnetic field in the region between the shim plates.

The runs were about equally divided between irradiations in which neutron polarizer field and sample polarizer field were parallel and those in which they were antiparallel. The state of polarization of the neutron beam transmitted by the sample was monitored throughout every sample irradiation by means of the "shim effect," G. Consider the case when neutron polarizer and sample polarizer are parallel. The field directions between all pairs of shims and the fields of the neutron polarizer, sample polarizer, and neutron analyzer were all adjusted to be in the same direction, and the counting rate, C_1, was taken. The three pairs

[13] D. J. Hughes and M. T. Burgy, Phys. Rev. **81**, 498 (1951).
[14] C. G. Shull, Phys. Rev. **81**, 626 (1951).

of shims c, d, and e were then adjusted so that a region of very nearly zero field was achieved in d. A thin sheet of unmagnetized iron was placed in this region of near zero field in the path of the beam, causing the beam to become depolarized.[15] The counting rate, C_2, was taken under these conditions.

The shim effect, G, is given by the expression

$$G=C_1/C_2=1+f_n f_n', \tag{4}$$

where f_n is the polarization produced by neutron polarizer alone, and f_n' is the neutron polarization produced by neutron analyzer alone. The shim effect is thus proportional to the polarization of the neutrons in the region between neutron polarizer and analyzer. For the antiparallel arrangement of neutron polarizer and sample polarizer the counting rate C_1 was taken with the fields of shims c, d, and e, and the field of neutron analyzer adjusted parallel to the sample polarizer field. For C_2, shim fields c, d, e were again adjusted to give a zero field region in which to depolarize the beam with the iron foil.

The shim effect is greater than 1 for the parallel field case and less than 1 for the antiparallel field case. $(G-1)$ will be symmetrical about 1, if the polarizations in the two cases have equal magnitude and opposite sign. The quantity $(G-1)$ was used as a relative measure of the polarization of the neutrons transmitted through the sample. Strictly speaking, the measured values of $(G-1)$ are proportional to the product of the neutron polarizations produced by polarizer alone and analyzer alone averaged over the continuous spectrum of neutrons emerging through 4.0 cm of polarizer iron plus 3.2 cm of analyzer iron, and weighted with the efficiency *vs* energy of the long BF_3 detector used. The polarization entering into the quantitative interpretation of the capture cross section data is the polarization detected by a $1/v$ detector averaged over the continuous spectrum emerging from the 4-cm thickness of polarizer. The measurement of this average polarization has been described.[12] About 80 percent to 95 percent of the continuous spectrum of neutrons were flipped. Under ideal conditions essentially all of the neutrons of approximately Maxwellian incident distribution filtered by a few cm of iron can be induced to make nonadiabatic transitions.[16] In our work the optimum magnetic field conditions for maximum efficiency of flipping were made somewhat difficult by the fact the the 11 000-oersted field at the neutron polarizer was only about 4 feet away from the oppositely directed 2350 to 10 500-oersted field of the sample polarizing magnet. The fraction of neutrons at the low and high energy ends of the spectrum was greater in our experiment than in the spectrum of reference 16 because the polarizing iron was thicker and the temperature of the incident Maxwell distribution of neutron velocities was higher.

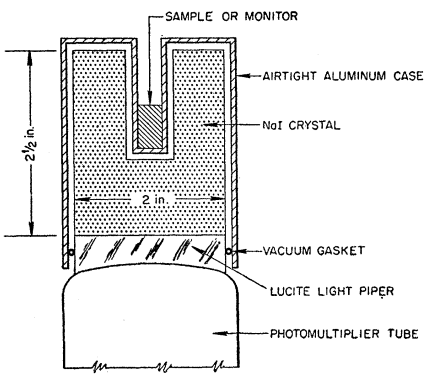

FIG. 3. Arrangement for measuring activities.

MEASUREMENTS OF ACTIVITIES

In order to avoid the necessity for making absolute measurements, a Mn metal monitor was used. This monitor was fixed in an accurately reproducible position to the outside of the Teflon box containing the sample salt, as shown in Fig. 2, and was exposed simultaneously with the salt. The monitor was machined from a cast ingot of Mn metal containing about 3 percent Cu. The Cu was added to make the material more easily machinable and less porous. Activity due to the 12.9-hour Cu^{64} was found to be unobservable in a series of test irradiations of 50 minutes made under the usual conditions of the experiment. The 4.3-minute Cu^{66} activity gave no contribution during the counting period since this short life-time activity died away during the period of about 50 minutes elapsing between the end of irradiation and the beginning of the counting. Both sample and monitor were found to have the accepted value for the half-life of 2.60 hours to less than 1 percent. The neutron beam passed through the monitor before impinging upon the sample.

The activity of the 2.6-hour Mn^{56} was measured by means of a NaI fluorescent crystal counting arrangement. The crystal was about 2-in. in diameter and $2\frac{1}{2}$-in. high. The samples were placed at the bottom of a reentrant "well" as shown in Fig. 3, to achieve greater efficiency. The walls of the sealed container surrounding the crystal were such that almost all of the pulses counted were due to three γ rays emitted by Mn^{56} and very few to β rays. The usual arrangement of fluorescent crystal, light piper, photomultiplier tube, amplifier, and scaling circuit was used.

The requirement on the counting system was simply that its over-all sensitivity remain constant over the 6-hour period during which the activities of a sample and monitor from a single irradiation were being measured. The sensitivity was monitored and adjusted by means of the 0.669-Mev 37-year Cs^{137} gamma ray. The observed integral and differential counting rate curves

[15] O. Halpern and T. Holstein, Phys. Rev. **59**, 960 (1941).

[16] Burgy, Hughes, Wallace, Heller, and Woolf, Phys. Rev. **80**, 953 (1950).

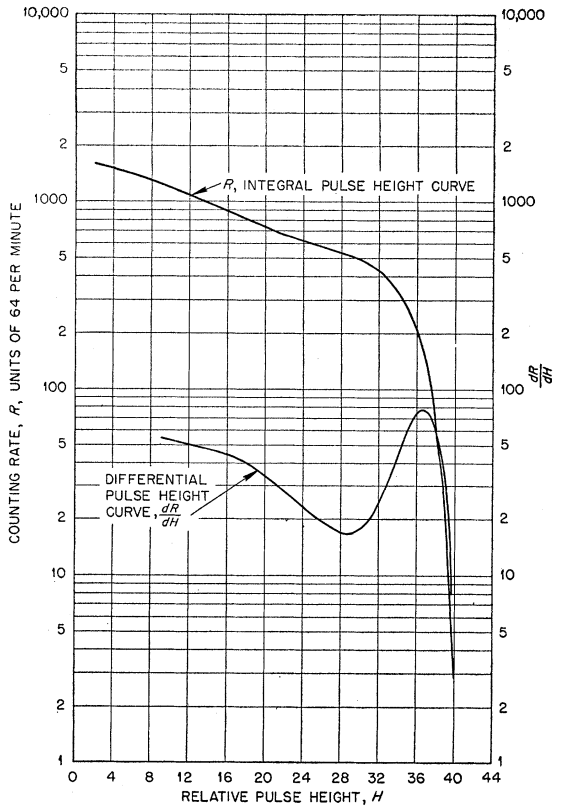

FIG. 4. Counting rate curves for Cs^{137}.

using Cs^{137} are shown in Fig. 4, in which the peak of the differential curve corresponds to 0.669 Mev. The gain of a photomultiplier tube is a very sensitive function of its applied voltage. To maintain constant sensitivity the voltage on the tube was adjusted about once per hour during the counting schedule to make the Cs^{137} peak of the differential curve occur at a fixed pulse height. In practice, one chooses the integral counting rate at a pulse-height setting corresponding to the peak and adjusts the voltage periodically to reproduce this counting rate.

The over-all gain of the system was actually tested with a much greater sensitivity than was used when counting the Mn activity. For Cs the pulse-height setting used was such that a small change in gain would cause a very large fractional change in counting rate. The pulse height used in counting the Mn activity was chosen to correspond to a rather flat portion of its integral counting rate *vs* pulse-height curve, thus making the Mn counting rate comparatively insensitive to gain changes. The Cs monitor count reproduced itself from hour to hour within 2 percent, or twice its statistical error. If this fluctuation were interpreted as being due to a true change in gain, then the Mn counting rate would have been affected by less than 0.05 percent. Differential counting rate curves were taken only for the purpose of checking and adjusting the equipment and finding optimum pulse-height settings for the integral counting rates taken during a run. For the irradiated samples all that was required was a number proportional to the saturated activity.

Counting rates of sample and monitor were taken alternately according to a planned schedule over a period of about six hours, starting 50 minutes after the end of the exposure. Positions of sample and monitor within the NaI crystal arrangement were made accurately reproducible. The relative orientations of sample and monitor obtaining during neutron irradiation were maintained during the activity measurements. This relative orientation was also kept the same from day to day. Initial counting rates for the sample were about 3000 counts per minute, and for the monitor were about 20 000 counts per minute. The NaI crystal background counting rate was about 100 counts per minute. For each run a total of about 3×10^5 counts were collected for the sample and about 10^6 counts for the monitor. Typical decay curves for the sample and monitor are shown in Fig. 5. The result of a single irradiation is given by the ratio of the saturated activity of the sample to that of the monitor. By this procedure all activity measurements of the sample were made relative to the activity of the monitor. Fluctuations in neutron beam intensity due to fluctuations in pile power were canceled out.

RESULTS AND ANALYSIS OF THE ACTIVATION MEASUREMENTS

Four types of irradiation were made. These are designated as I, II, III, and IV in the first column of Table I. The conditions listed in this column are the final temperature and magnetic field values of the sample, and the relative directions of sample-polarizing and neutron-polarizing fields. A comparison of the results of I and II should show the full effect of relative particle orientation. Under the temperature and field conditions of III and IV, the sample should have the same electron polarization but much less nuclear polarization than under the conditions of I and II. The relative change in sample activity exhibited by III and IV should be

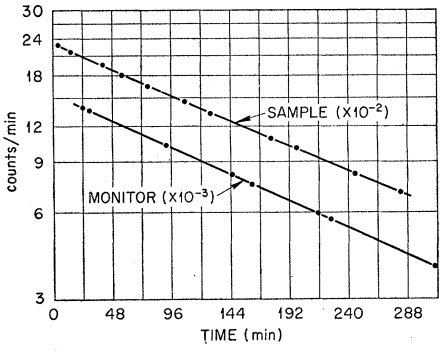

FIG. 5. Typical decay curves of sample and monitor.

TABLE I. Results and analyses of activation measurements.

Experimental conditions (1.)	Run number (2.)	$R=\frac{A(S)}{A(M)}$ (3.)	Standard deviation of R, $\sigma(R)$ (4.)	Relative error in R, % $100\times\sigma(R)/R$ (5.)	Weighting factor, $w=[1/\sigma(R)]^2$ (6.)	Flipping efficiency factor; q (7.)
I	1	0.1443	8.66×10^{-4}	0.60	1.334×10^{6}	
$T=0.20$°K	4	0.1446	7.95×10^{-4}	0.55	1.581×10^{6}	
$H=2350$	5	0.1439	7.34×10^{-4}	0.51	1.857×10^{6}	
oersteds	8	0.1450	7.83×10^{-4}	0.54	1.631×10^{6}	
Fields	10	0.1439	14.82×10^{-4}	1.03	0.455×10^{6}	
parallel	11	0.1445	9.97×10^{-4}	0.69	1.006×10^{6}	
	14	0.1431	15.03×10^{-4}	1.05	0.443×10^{6}	
	16	0.1430	7.44×10^{-4}	0.52	1.809×10^{6}	
	34	0.1433	7.60×10^{-4}	0.53	1.733×10^{6}	
	36	0.1434	12.76×10^{-4}	0.89	0.614×10^{6}	
II	2	0.1519	14.28×10^{-4}	0.94	0.490×10^{6}	0.83
$T=0.20$°K	3	0.1499	9.14×10^{-4}	0.61	1.196×10^{6}	0.91
$H=2350$	6	0.1489	9.08×10^{-4}	0.61	1.212×10^{6}	0.86
oersteds	7	0.1471	7.65×10^{-4}	0.52	1.709×10^{6}	0.77
Fields	9	0.1488	8.04×10^{-4}	0.54	1.549×10^{6}	0.91
antiparallel	12	0.1482	10.82×10^{-4}	0.73	0.862×10^{6}	0.84
	13	0.1499	10.34×10^{-4}	0.69	0.935×10^{6}	0.85
	15	0.1498	13.33×10^{-4}	0.89	0.563×10^{6}	0.86
	17	0.1485	21.24×10^{-4}	1.43	0.122×10^{6}	0.84
	18	0.1458	16.18×10^{-4}	1.11	0.382×10^{6}	0.85
	35	0.1487	8.48×10^{-4}	0.57	1.392×10^{6}	0.83
	37	0.1491	7.01×10^{-4}	0.47	2.036×10^{6}	0.85
III	19	0.1463	7.31×10^{-4}	0.50	1.869×10^{6}	
$T=1.2$°K	21	0.1462	12.57×10^{-4}	0.86	0.633×10^{6}	
$H=10\,500$	23	0.1451	6.53×10^{-4}	0.45	2.345×10^{6}	
oersteds	25	0.1459	6.57×10^{-4}	0.45	2.320×10^{6}	
Fields	27	0.1478	13.60×10^{-4}	0.92	0.541×10^{6}	
parallel	30	0.1460	6.13×10^{-4}	0.42	2.660×10^{6}	
IV	20	0.1478	10.64×10^{-4}	0.72	0.883×10^{6}	0.71
$T=1.2$°K	22	0.1460	7.45×10^{-4}	0.51	1.804×10^{6}	0.81
$H=10\,500$	24	0.1452	7.12×10^{-4}	0.49	1.975×10^{6}	0.93
oersteds	26	0.1474	6.63×10^{-4}	0.45	2.273×10^{6}	0.92
Fields	28	0.1477	7.98×10^{-4}	0.54	1.572×10^{6}	0.92
antiparallel	29	0.1484	7.72×10^{-4}	0.52	1.679×10^{6}	0.84
	31	0.1471	7.36×10^{-4}	0.50	1.848×10^{6}	0.96
	32	0.1473	13.55×10^{-4}	0.92	0.545×10^{6}	0.99
	33	0.1454	9.60×10^{-4}	0.66	1.086×10^{6}	0.92

about $\frac{1}{6}$ of the change shown by comparison of I and II.

The total number of counts, z_i, accumulated during the time interval $(t_i-t_i^*)$ were recorded at various times in the course of the decay of the activity of sample and monitor. The intervals $(t_i-t_i^*)$ varied from 5 to 30 minutes, depending upon whether the sample or monitor was being counted and upon the elapsed time between the end of irradiation and time of counting. The counts, z_i, are given by the expression

$$z_i=A[\exp(-\lambda t_i)-\exp(-\lambda t_i^*)]+B(t_i-t_i^*). \quad (5)$$

In (5), B is the background counting rate of the counting system, A is a number proportional to the number of activated atoms at $t=0$. It was assumed that λ, the decay constant of Mn^{56} and the times t_i and t_i^* were known with negligible error. The expression (5) can then be written

$$z_i=Ax_i+By_i, \quad (6)$$

in which z_i, x_i, and y_i are measured quantities. For a typical activation, the number of z_i's or the number of intervals, n, for each of which a total count was taken, was about ten. Each of the z_i for the sample consisted of roughly 25 000 counts, and for the monitor of 75 000 counts.

All of the activity data were analyzed by conventional statistical procedures. The relative activity of the sample, as determined by a single irradiation is given by

$$R=A(S)/A(M), \quad (7)$$

in which $A(S)$ and $A(M)$ are the most probable values respectively of A for the sample and monitor. The values of R are given in column 3 of Table I, and the standard deviation of each R is given in column 4. Column 5 gives the relative standard deviation of each R in percent. In calculating the average value $\bar{R}$, of the measured R's of a single group of runs, the individual R's were weighted by the reciprocal of the corresponding variance of each R. These weighting factors, w, are given in column 6.

The values of $\bar{R}$ for the four types of irradiation are given in column 2 of Table II. The variances in $\bar{R}$ are given in column 3 and the relative standard deviations in percent are given in column 4. The values of the $\bar{R}$'s are: (I) 0.1440 ± 0.0002; (II) 0.1488 ± 0.0004; (III) 0.1459 ± 0.0004; (IV) 0.1469 ± 0.0004. The relative difference in activity of the sample with change in relative orientation of incident and bombarded particles is given by

$$D=(\bar{R}_a-\bar{R}_p)/\bar{R}_p, \quad (8)$$

in which $\bar{R}_a$ is the average of R for a group of runs taken under a given set of final magnetic field and temperature conditions for the sample, with the sample polarizing field and neutron polarizing field antiparallel, and $\bar{R}_p$ is the average value of R for another group of runs taken under the same final sample temperature and field conditions with the two polarizing fields parallel. The values of D in percent are given in column 5. The standard deviations of the D's are given in column 6. The final results state, then, that at 0.20°, the sample activity with fields antiparallel was (3.38 ± 0.34) percent greater than the activity with fields parallel. At 1.20°K, with the sample possessing

TABLE II. Summary of statistical averages of the activity data.

(1)	$\bar{R}$ (2)	$[\sigma(\bar{R})]^2$ (3)	$\frac{100\sigma(\bar{R})}{\bar{R}}$ (4)	$(\bar{R}_a-\bar{R}_p)/\bar{R}_p$ D,% (5)	$\sigma(D)$ (6)	t (7)
I $T=0.20°$, $H=2350$, parallel	0.1440	4.87×10^{-8}	0.153			
				3.38	0.34	9.3
II $T=0.20°$, $H=2350$, antiparallel	0.1488	19.30×10^{-8}	0.295			
III $T=1.2°$, $H=10\,500$, parallel	0.1459	15.03×10^{-8}	0.266			
				0.66	0.33	1.7
IV $T=1.2°$ $H=10\,500$, antiparallel	0.1469	14.03×10^{-8}	0.255			

within 1 percent, the same electron polarization and much less nuclear polarization than at the lower temperature, the sample activity with fields antiparallel was (0.66±0.33) percent greater than with the fields parallel.

A parameter which is useful in discussing the reliability of the final results is the ratio, t, of the value of $(\bar{R}_a-\bar{R}_p)$ to the standard deviation of this difference.[17]

$$t=\frac{\bar{R}_a-\bar{R}_p}{\sigma(\bar{R}_a-\bar{R}_p)}. \tag{9}$$

The values of t are given in column 7 of Table II. For the runs at 0.20°, 2350 oersteds, the value of t is about 9. If one assumes, for purpose of illustration, that the two R's at 0.20° for parallel and antiparallel orientations came from the same population, then it can be demonstrated by standard statistical procedures that the probability of obtaining a value of 9 or greater for t from 22 runs, is extremely unlikely. Similarly, for the data of 1.2° and 10 500 oersteds, the probability is about 1 in 16 that a value of $t=1.7$ or greater will be obtained from 15 runs belonging to the same population, and corresponding to the same direction of effect as in 0.20° group. It seems, then, that the 3.38 percent change in activity obtained at 0.20°, and 2350 oersteds is, statistically at least, a "real" one with a very high degree of probability, whereas the 0.66 percent change in activity with relative orientation observed at 1.2° and 10 500 oersteds has a very much lower probability of being statistically "real".

Some aspects of the data are illustrated graphically in Fig. 6, a histogram of the R_j values obtained with the four different experimental arrangements. The ordinate gives the relative number of runs, weighted with the weighting factor w_j. The width of the histogram interval is the value of $\sigma(R)$ averaged over all the runs of the four types. The areas enclosed by the histograms for the two types of irradiation at a single temperature were made equal. The average values $\bar{R}$ for each of the four types are indicated by the arrows along the axes of abscissas. The number above each histogram ordinate is the actual number of runs whose R values fall in that interval.

Column 7 of Table I gives values of a flipping efficiency factor q, derived from the shim effect values measured during each irradiation. The value q is the fraction of the neutrons which were induced to make the required nonadiabatic transition in the antiparallel runs. These q values can be used to correct the values of D to the values of D which would have been obtained if all the neutrons had been flipped. When this correction is made the value $D=3.38$ percent is increased to the value $D=4.0$ percent.

The change in nuclear cross section on polarization is to a good approximation inversely proportional to

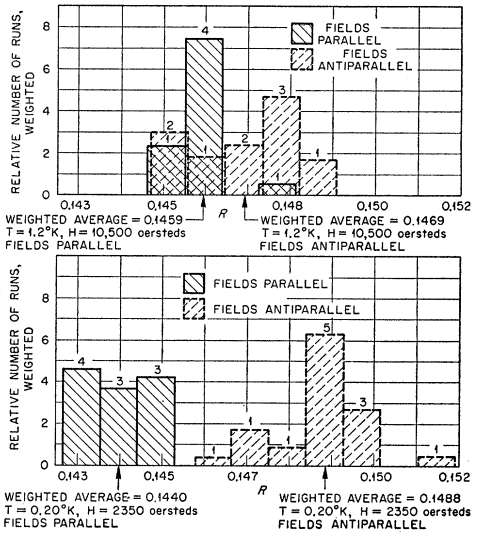

FIG. 6. Histogram of the experimental results. The ordinate is the relative frequency with which the relative activity given by the abscissa was obtained. The width of the interval is the average standard deviation of the activity.

the temperature at constant electronic magnetic moment. Although the fractional error on the 0.66 percent is large, the ratio $3.38/0.66\approx 5.1$ of the cross section changes observed at 0.20°K and 1.20°K, respectively, is close to the inverse ratio of the corresponding temperatures, $1.2°/0.20°=6.0$. This good agreement may be accidental but it does serve to emphasize that with constant neutron and electron polarizations the change in cross section with the two types of relative orientation of the particles decreased as the temperature increased in accordance with the theory. The average value $\frac{1}{2}(\bar{R}_p+\bar{R}_a)=0.1468$ of all runs at 0.20° including both parallel and antiparallel is very close to the value 0.1465 for the same average for all runs at 1.20°K. Both of these averages have been corrected for flipping efficiency. Throughout the entire experiment the distribution of repeated measurements of the same quantity compared very favorably with the distribution expected from a consideration of the counting statistics of a single measurement.

The electronic magnetic moment was measured immediately before and after each irradiation. The nuclear polarization varied slightly from run to run. For the 0.20°K measurements, the variations occurred during the irradiations because of the ~100 erg/min heat leak to the sample, and from day to day because of small variations in the initial and final fields and initial temperatures used. Variations in the field and temperature used for the 1.20°K measurements were extremely small. The calculated nuclear polarization for all 0.20°K runs fell in the range $f_N=0.16\pm0.01$, and the 1.20°K runs in the range $f_N=0.0265\pm0.0009$. Each of the sample to monitor activity ratios given in Table I required one day of work in preparation and

[17] A. M. Mood, *Introduction to Theory of Statistics* (McGraw-Hill Book Company, New York, 1950), p. 225.

measurement. The time order of the runs is given by the run numbers in the second column. Runs in groups I and II were made on alternate days insofar as possible, as were those of III and IV. The 1.20°K runs of III and IV took place after run number 18 of II, after which 4 runs (34–37), again at 0.20°K were performed. The purpose of this time order was to check on any possible changes with time of the sample. No changes are apparent, however. All of the runs of Table I were taken with a single sample

The weight of the sample was recorded after each run to check on possible changes due to dehydration, exchange of hydrogen for deuterium, and mechanical losses. Over the period of the two months during which the data were taken the sample weight changed by about 0.6 percent, and the deuteration decreased less than one percent.

TRANSMISSION EXPERIMENTS

The statistical analysis of the activation data suggests very strongly that the change in activity of the Mn as a function of relative orientation of neutrons and nuclei is real. The question can be raised as to whether or not the change in activity is correctly attributed to a change in the capture cross section of Mn. If, for some reason, there were a change of average path length through the sample of the scattered neutrons in going from the parallel to the antiparallel arrangement, the result would be a change in the activity of the sample, or an apparent change in capture cross section.

In order to determine if a change of total cross section accompanied the change of activity of the sample, a a series of transmission measurements for polarized incident neutrons were taken with the sample under the conditions described in Table III. The neutron polarizer magnetic field and the sample field were parallel for runs 2 and 4. The neutron detector slit, 1 inch wide and 2 inches high, was located about six feet behind the sample for the transmission measurements.

Comparison of the transmission in runs 1 and 2 should show the effect on the total cross section of the magnetic scattering due to electron polarization. Comparison of runs 3 and 4 should show the effect of nuclear polarization on the transmission. Comparison of runs 1 and 3 should show the effect of a change in sample temperature from 1.2°K to 0.2°K upon the transmission.

TABLE III. Conditions of sample during transmission measurements.

Run	Temperature, °K	Magnetic field, oersteds
1	1.20	0
2	1.20	16 400
3	0.12	0
4	0.20	2350

The transmission of the sample was found to be the same for all of the conditions listed in Table III, within the statistical error of the transmission measurements, which was ± 0.2 percent showing that any changes in transmission due to the above mentioned causes were quite negligible compared to the observed change of 3.38 percent in sample activity with relative orientation of neutrons and nuclei. The transmission experiments show that the number of transmitted neutrons (and the number of scattered neutrons) remained unchanged with change of electron and neutron polarization. Any possible change of average path length must, therefore, take place without change in the number of scattered neutrons. Such a change in angular distribution of scattered neutrons with electron or neutron polarization without a corresponding change in the total cross section seems difficult to understand.

The transmission of the sample was 60.9 percent. This low value of the transmission is caused by scattering from the many atoms other than Mn making up the $MnSO_4 \cdot (ND_4)_2SO_4 \cdot 6D_2O$. Only about 2.5 percent of the neutrons incident upon the sample are captured by the Mn. A change of 3.38 percent in the number of neutrons captured by Mn would change the sample transmission by about 0.1 percent. Such a change is about equal to the statistical error of the transmission measurements and could not be detected. Any change in the scattering cross section of the Mn nucleus due to its polarization would be even less noticeable in the transmission measurements, since the nuclear scattering cross section of Mn is much smaller than its capture cross section for slow neutrons.

DEPOLARIZATION OF THE NEUTRON BEAM BY COLLISIONS

The polarization of the neutron beam can be reduced by scattering collisions of the neutrons, with those constituents of the Dewar flask walls, the monitor, the sample itself, and other materials in the path of the beam, which have nonzero nuclear spins. We have calculated an extreme upper limit to the extent of this depolarization by following the average polarization through several collisions of those neutrons passing through the sample, on the basis of very pessimistic assumptions. Of those neutrons scattered by materials other than the sample, we have assumed that fully one-half pass through the sample, and that the average path length in the sample of all scattered neutrons is the same as for the unscattered neutrons. In the case of isotopes for which the spin dependent amplitudes were not known, the assumption was made that all those neutrons scattered incoherently were depolarized. For the case of H and D the published values of the spin dependent scattering amplitudes were used to calculate an upper limit of the probability[18] for the inversion of the spin during a scattering process.

[18] O. Halpern, Phys. Rev. 88, 232 (1952).

The most important factor in the depolarization is the initial scattering by the deuterons of the sample. Successive collisions with sample, monitor, and walls reduce the polarization an additional small amount. The result of the calculation is that the average polarization of those neutrons captured by the sample is, at the very least, about 85 percent of the initial neutron polarization.

INTERPRETATION OF THE DATA IN TERMS OF THE NUCLEAR PROPERTIES OF Mn

The interpretation of our data will depend upon the sign of the hyperfine structure coupling constant, A, in the expression $A\mathbf{I}\cdot\mathbf{S}$ for the energy of interaction of the nuclear angular momentum of quantum number, I, and the angular momentum of the ionic electrons of quantum number, S. The sign of A has been found to be negative,[10] so that the nuclear spin and electronic spin of Mn are parallel in the energy levels of greatest population. The interpretation depends also on the direction of the spins of the neutrons emerging from the polarizing block of iron. The direction of the spins of the neutrons emerging from the polarizing iron is known from theory and experiment[19] to be antiparallel to the direction of the magnetic field applied to the polarizing block. The result of these two statements is that the spins of the incident neutrons and the bombarded Mn nuclei are parallel when the magnetic fields applied to the neutron polarizing block and the sample are parallel.

When this experiment was being planned, the compound nucleus, Mn^{56}, was thought to have the resonance levels[20] with nuclear parameters[21] shown in Table IV. It was thought that the size of effect to be observed at thermal neutron energies would be determined largely by the 345 ev level for which $J=3$. On this basis, according to (3a) and (3b), the ratio, $\bar{R}_p/\bar{R}_a$, of the activity for magnetic fields (spins) parallel to the activity for fields (spins) antiparallel should be about 1.068. Actually, this ratio of $\bar{R}_p/\bar{R}_a$ was measured to be 0.966, and becomes 0.96 when corrected for neutron flipping efficiency.

If both the 345 ev and the 2400 ev levels influence the thermal capture cross section, then the cross section is given by the sum of the two expressions on the right side of relations (3a) and (3b), in which sum σ_1 is the capture cross section due to the 345 ev level only, σ_2 is the capture cross section due to the 2400 ev level only. The ordinary cross section for no polarization is $\sigma_0=\sigma_1+\sigma_2$. σ_1 and σ_2 each obey the one level Breit-Wigner relation for the capture cross section. The upper level for which $J=2$ affects the spin dependence of the capture cross section in the opposite direction to the lower level. If one uses the sum of (3a) and (3b) with the Breit-Wigner expression and the level parameters given in Table IV, considering Γ_γ of the upper level as the unknown to be determined by the data, then Γ_γ must become unusually large in order that the upper level over-ride the effect of the lower and give the observed $\sigma/\sigma_0=0.96$ at thermal energies.

TABLE IV. Breit-Wigner parameters for nuclear levels of Mn^{56}.

E_0, Neutron energy ev	Γ_n, Neutron width, ev	Γ_γ, γ-ray width, ev	J, angular momentum
345	21.6	0.6	3
2400	304.	about 1 ev	2

Recent measurements[22] of the total cross section of Mn have brought to light another resonance level at about 1000 ev. The Breit-Wigner parameters of this level are not known yet. There is also the possibility of an unknown resonance level at negative energy influencing our results. It seems, then, that as of this date, there is not enough information available on the nuclear energy level system of Mn^{56} to state definitely whether or not the nuclear properties and the hyperfine structure magnetic resonance measurements are consistent with our observation that those thermal neutrons whose spins are antiparallel to the spins of Mn^{55} nuclei have a greater probability of being captured than those whose spins are parallel.

Note added in proof.—Recent mechanical velocity selector measurements of Bollinger, Palmer, and Dahlberg (Bulletin of the American Physical Society, Vol. 29, No. 4, 1954, Washington meeting) show neutron resonances in Mn^{55} at 337, 1080, and 2360 ev, to which they assign the J values of 2, 3, and 3, respectively.

ACKNOWLEDGMENTS

We are very much indebted to Drs. M. E. Rose, J. M. Jauch, A. Simon, M. Hamermesh, G. Trammell, and T. A. Welton for discussions of the theoretical physical aspects of this work and to Drs. J. Moshman, C. L. Perry, and A. S. Householder for help with the statistical aspects. The mass spectrographic analyses of the sample were made by Dr. R. Baldock and his group. Miss T. Arnette assisted in taking the data. Mr. J. B. Capehart assisted with the cryogenic aspects of the work.

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[22] D. J. Hughes, Brookhaven National Laboratory (private communication).