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CF-55-5-126

Subject Category: PHYSICS

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UNITED STATES ATOMIC ENERGY COMMISSION

DIRECT POLARIZATION OF In^{115} NUCLEI:
J VALUE FOR 1.456 eV RESONANCE

By
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June 1955

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Work performed under Contract No. W-7405-Eng-26

OAK RIDGE NATIONAL LABORATORY

Operated by

CARBIDE AND CARBON CHEMICALS COMPANY

A Division of Union Carbide and Carbon Corporation

Post Office Box P

Oak Ridge, Tennessee

ACKNOWLEDGEMENTS

Dr. C. P. Stanford contributed materially to the neutron aspects of Experiments I, II, and III, and Mr. E. Soltysik of Indiana University contributed his efforts during a summer visit. The authors are indebted to Dr. M. E. Rose for many stimulating discussions, especially with regard to the "Majorana flipping" of neutrons, and to Dr. E. O. Wollan for several interesting suggestions regarding neutron diffraction. The authors wish to thank Dr. E. C. Smith for his kindness in remeasuring the transmission of a laboratory standard BF_3 neutron absorption cell, and Dr. H. H. Landon for permission to quote his results prior to publication. The authors would like to thank Mr. J. B. Capehart for technical assistance, Mr. R. H. Ward and Mr. C. R. Hill of the Physics Division Shop for their many contributions.

TABLE OF CONTENTS

CHAPTER	PAGE
I. NUCLEAR POLARIZATION	1
Introduction	1
Methods Proposed for Nuclear Polarization	2
Methods of Detection of the Polarization	6
Utilization of Nuclear Polarization	8
Purposes of the Present Investigation	11
II. THE BRUTE FORCE METHOD, USING NEUTRONS	12
Choice of In ¹¹⁵	17
Choice of Cooling Salt	23
III. CRYOGENIC DESIGN OF THE EXPERIMENTS	31
Introduction	31
Heats to be Absorbed by the Cooling Salt	32
Previous Development of the Demagnetization Apparatus	33
Developments for Present Experiments	37
Transient Heat Inputs	43
Steady Heat Inputs	51
Effects of Heat Input	54
General Conclusions	63
IV. NUCLEAR DESIGN OF THE EXPERIMENTS	65
Thickness of the In Sample	65
Neutron Polarization Methods	67

CHAPTER	PAGE
Neutron Flipping Methods	69
Neutron Depolarization	74
Neutron Heating in Various Arrangements	75
Double Crystal Spectrometer-Polarizer	79
V. AUXILIARY AND ASSOCIATED EQUIPMENT	86
Cryogenic Equipment	86
Nuclear Equipment	90
VI. DESCRIPTION OF EXPERIMENTS	94
Experiments I	94
Experiments II	95
Experiments III	96
Experiments IV	99
VII. DISCUSSION OF RESULTS	107
Direction of Effects	107
Magnitude of Effect in Experiments IV	114
Possibilities for Future Development	117
BIBLIOGRAPHY	119

CHAPTER I

NUCLEAR POLARIZATION

CF-55-5-126

Introduction

The investigation reported here is concerned with an experimental attempt to produce and observe a macroscopic magnetization of the nuclear magnetic moments of indium nuclei in the direction of an externally applied magnetic field. Such a magnetization has come to be called a nuclear polarization, in contrast to a nuclear alignment in which no net magnetization is produced, the magnetic moments being oriented along a spatial direction, but with no preferred sense. As will be seen, a direct or "brute force" nuclear polarization can be produced by cooling the nuclear sample to a very low temperature ($\ll 1^\circ\text{K}$) and subjecting it to a strong external magnetic field. This cooling was accomplished in the present instance by thermally connecting the nuclear sample to a paramagnetic salt (ferric ammonium alum) which was cooled by adiabatic demagnetization. The polarization was then obtained by the application of the external field. The present investigation was carried out to demonstrate a direct nuclear polarization; to determine the total angular momentum of the compound nucleus formed when the nucleus In^{115} captures a thermal energy neutron, and to develop some of the techniques necessary to the performance of a nuclear adiabatic demagnetization.

In 1934, Gorter, in a discussion following a paper of P. Debye¹, made the first suggestion that the polarization of nuclear magnetic moments could give a method for obtaining extremely low temperatures by nuclear demagnetization. The same suggestion was independently made by Kürti and Simon² in 1935, and discussed in some detail by Simon³ and by Kürti⁴. The method of nuclear polarization which was envisaged in these discussions was quite simple; it consisted of applying an external magnetic field directly to the nuclear magnetic moments at as low a temperature as could be reached. This method has since come to be called the "brute force" method and the term will be applied here. At the time of these discussions, no suggestions for the use of polarized nuclei to study properties of nuclei were made.

Methods Proposed for Nuclear Polarization

Since the discussions mentioned above several suggestions have been made for polarizing nuclei in solids in which restrictions of one kind or another as to the species of nuclei to be polarized or to the nuclear surroundings have been accepted.

¹P. Debye, Phys. Zeits. 35, 923 (1934).

²N. Kürti and F. E. Simon, Proc. Roy. Soc. 149, 152 (1935).

³F. E. Simon, Comptes Rendues du Congrès sur le Magnétisme, Strasbourg, V. 3, 1. (1939).

⁴N. Kürti, Les Phénomènes Cryomagnétiques, Collège du France, 16 Nov. 1948, p. 29.

Rose - Gorter Method

In 1948, Rose⁵ and Gorter⁶ independently proposed the polarization of nuclei by utilizing materials in which the hamiltonian of the system could essentially be written as the sum of two terms, these being (1) the interaction of an external field with an unpaired electron spin, $g\beta\tilde{H}\cdot\tilde{S}$, and (2) a h.f.s. (hyperfine structure) coupling, $A\tilde{I}\cdot\tilde{S}$, between the nuclear spin operator $\tilde{I}$ and the electron spin operator $\tilde{S}$, A being the coupling constant. In fields such that the first term is large compared to the second (Paschen-Back region), the electron spins are oriented relative to the external field H and these electrons may be said to produce at the position of their nuclei an effective magnetic field of the order of $10^5 - 10^6$ gauss for reasonable values of A , say $0.01 - 0.1 \text{ cm}^{-1}$. In this method, the magnetic fields required are quite modest, since unpaired electron spins may be obtained only in materials with unfilled electron shells, and such materials (e.g., paramagnetic salts of the transition, rare-earth, or transuranic groups) are often also suitable for adiabatic demagnetization, or could be made so by dilution or mixing with other (perhaps isomorphous) salts. This implies that they may be cooled to very low temperatures, at which point an external field of perhaps a thousand gauss should be sufficient for a number of substances to essentially saturate the electron spin system, giving large nuclear polarizations. Ex-

⁵M. E. Rose, AECD 2119 (July, 1948); Nucleonics 3, No. 6, 23 (1948); Phys. Rev. 75, 213 (1949).

⁶C. J. Gorter, Physica 14, 504 (1948).

periments using this method have been carried out at Oak Ridge National Laboratory^{7,8} and elsewhere⁹. The method, however, does not permit decoupling the nuclei thermally from the system used in their polarization; this implies that nuclear demagnetizations cannot be accomplished by this means.

Overhauser's Method

In 1953, A. W. Overhauser¹⁰ proposed a method suitable for obtaining polarization of nuclei in metals. The method takes advantage of electron spin resonance of the conduction electrons in a magnetic field. If this system is saturated by resonance radiation, the nuclei behave essentially as if they possessed the same gyromagnetic ratio as electrons, and the polarization may become quite large if the relaxation times involved are of proper magnitudes. The method suffers from the requirement of rather large power in the microwave region, and the dissipation of this power, probably by boiling of a liquid helium bath. The metal would also have to be finely divided, the particles probably not exceeding about one micron in diameter. Experiments at room temperature¹¹ have demonstrated the method. However, this method has not been investigated as yet from

⁷Bernstein, Roberts, Stanford, Dabbs, and Stephenson, Phys. Rev. 94, 1243 (1954).

⁸Roberts, Bernstein, Dabbs, and Stanford, Phys. Rev. 95, 105 (1954).

⁹Ambler, Grace, Halban, Krti, Durand, Johnson, and Lemmer, Phil. Mag. 44, 216 (1953).

¹⁰A. W. Overhauser, Phys. Rev. 92, 411 (1953).

¹¹T. R. Carver and C. P. Slichter, Phys. Rev. 92, 212 (1953).

the standpoint of performing a nuclear demagnetization from a system in this way; this is, therefore, a question which should be investigated. It should also be noted that, obviously, a large amount of development work will be required to realize large polarizations in this manner. We shall, therefore, omit this method from further consideration.

Several investigators^{12,13,14} have recently extended the suggestion of Overhauser to the case of paramagnetic materials. They show that in this case, the nuclear polarization finally obtained is independent of the relaxation times involved for a given electron saturation although the approach to the polarization would, of course, be governed by these times. Here, long relaxation times are desirable to reduce the amount of microwave power required. As before, no suitable method of thermally decoupling the nuclear system from the large specific heat associated with the paramagnetic electron system exists, and this prevents use of this method of polarization as a first step in a nuclear demagnetization.

At present, the "brute force" method would appear to be the most feasible approach to the development of an experiment leading to a nuclear demagnetization.

The "Brute Force" Method

If a system of nuclei with magnetic moments $\mu = g_N \beta_N I$ is placed

¹²J. Korringa, Phys. Rev. 94, 1388 (1954) and references cited there.

¹³Brovetto and Cini, Nuovo Cimento 11, 618 (1954).

¹⁴Brovetto and Ferroni, Nuovo Cimento 12, 90 (1954).

in a magnetic field H , and we neglect the interactions among nuclei and assume there is no h.f.s. coupling, the hamiltonian of the system is given by

$$\mathcal{H} = - g_N \beta_N \tilde{H} \cdot \tilde{I} \quad (1)$$

where g_N and β_N are the magnetogyric ratio and the nuclear magneton, respectively. If the nuclei are characterized by a spin angular momentum $\hbar I$ where $2\pi\hbar$ is Planck's constant, there will occur the well-known Zeeman splitting into $2I + 1$ energy levels with energies $-m_I g_N \beta_N H$ in the magnetic field, where m_I is the magnetic quantum number, ranging from $-I$ to $+I$. If then the system is allowed to come to thermal equilibrium at some absolute temperature T , a Boltzmann distribution of population over the $2I + 1$ energy levels will be found. The excess population in the lower energy levels corresponds to a nuclear polarization. The "brute force" method involves increasing this inequality of populations by the use of a large magnetic field applied at a very low absolute temperature.

Methods of Detection of the Polarization

Assuming that a sample of nuclei whose spins are polarized has been obtained in a solid, one must consider the means by which one might detect or measure the polarization achieved; and perhaps in so doing, one might inquire as to what additional information may be obtained in this process.

Spin Dependent Nuclear Interactions

Since nuclear polarizations of the type envisaged here imply, by

definition, inequalities in the population of the various spin states of the nuclei, an immediately obvious idea is to attempt to utilize the spin-dependence of nuclear interactions as a detection tool. One might, for example, bombard a polarized target with a beam of polarized particles and observe the change in capture or scattering cross sections with change in the predominant relative spin orientations of the beam particles and the nuclei of the target. Rose¹⁵ has suggested the use of neutron beams, polarized by magnetic interaction with ferromagnetic materials as a useful means, and two such experiments^{7,8} have been carried out at the Oak Ridge National Laboratory. The present work represents a third such experiment.

Nuclear Magnetic Resonance Absorption

Another possible detection method for polarized nuclei is to use nuclear magnetic resonance absorption. At ordinary temperatures, the absorption method depends on the extremely small amount of nuclear polarization present, for the net absorption of energy at the Larmor frequency is proportional to the differences in populations of the levels between which transitions are induced by the oscillating field. As the nuclear polarization is increased, the amount of absorption increases also. Thus a very sensitive detector is available. The drawbacks of this method for the present experiments are that (a) the application of nuclear magnetic

¹⁵M. E. Rose, AECD 2183 (July, 1948); Nucleonics 3, No. 6, 23 (1948); Phys. Rev. 75, 213 (1949).

resonance to metals below 1°K would present difficulties with respect to eddy current heating, and (b) relaxation time effects below 1°K would have to be carefully studied. The use of a metal is quite desirable for the present experiments because of the relatively large thermal conductivity at low temperatures, as well as the very reasonable nuclear relaxation times in metals.

Radioactive Decay

Although the method of using radioactive decay (anisotropy of γ -ray emission, for example) is primarily of interest in nuclear alignment¹⁶ as contrasted with nuclear polarization, it nevertheless, in some cases, serves the need of an indicator of the degree of polarization obtained and has been utilized in at least one instance⁹. The method has the further possibility of yielding rough measurements of the nuclear magnetic moments of radioactive nuclei. The major disadvantage lies in the fact that the same radioactivity which enables a measurement also heats the sample. This limits intensities and counting rates severely, so that statistical errors may become quite important.

Utilization of Nuclear Polarization

A very important question is concerned with the utilization of

¹⁶Simon, Rose and Jauch, Phys. Rev. 84, 1155 (1951). B. Bleaney, Proc. Phys. Soc. (London) A 64, 315 (1951); Phil. Mag. 42, 441 (1951).

nuclear polarization as a tool of research. The answers to this question have already been touched on above either implicitly or explicitly; but a brief statement of these answers is in order here.

Determination of Nuclear Properties

It is clear from the discussion above that a very desirable tool is available in polarized target nuclei for the study of neutron capture and scattering resonances. The most desirable situation would be, of course, to have a target of highly polarized nuclei, and to bombard such a target with a beam of highly polarized neutrons of easily and continuously variable energy; one could then take each resonance in turn, and by merely observing the direction of the change in transmission with, say, neutron spin direction, determine the angular momentum of the compound nucleus for each resonance. Such an experimental program would be a matter of great interest. Unfortunately, the only simple means of polarizing neutrons are at present restricted to the thermal neutron region. Clearly, one could produce polarized resonance neutrons by means of another sample of polarized nuclei, allowing these neutrons to impinge upon the second sample, and perhaps reversing the neutron spin direction or depolarizing the neutrons in the intervening space at will. This is clearly a formidable task. Another possibility would be to produce polarized neutrons (or protons) by means of nuclear reactions, as several groups of workers have recently done¹⁷; here, however, the polarization of the nucleons is energy

¹⁷Meier, Scherrer, and Trumpy, *Helv. Phys. Acta* 27, 577 (1954) and references cited there.

dependent, and intensity considerations will make the work difficult.

Temperature Measurement

The dependence of nuclear polarization on temperature is a strong one; i.e., in the "brute force" method, it is as $1/T$ for small polarizations, and for other methods may go as $1/T^2$. This immediately suggests that, given suitable detection means, the polarization may serve as a quite useful thermometer in the very low temperature region well below 1°K . Thus far, temperature measurements below 0.1°K are quite unsatisfactory, and nuclear orientation methods may provide the only satisfactory solution.

Nuclear Demagnetization

If one has a sample of polarized nuclei which may be thermally isolated from its surroundings, and if the heat input to the sample can be made quite small³, say 1 erg/min, it becomes feasible to carry out a nuclear adiabatic demagnetization analogous in all respects to the presently used adiabatic demagnetization of paramagnetic salts. It is expected that, generally, temperatures less than 10^{-4}°K would be obtainable in this manner, because the interactions between neighboring nuclei are very small. Many interesting investigations as to the behavior of collections of nuclei could be carried out if such experiments were made feasible. For example, one might expect that collections of nuclei would become either ferromagnetic or antiferromagnetic at sufficiently low temperatures. Studies of such behavior might lead to interesting conclusions about long-range

nuclear interactions and effects of atomic electrons on such interactions¹⁸; in short, a whole new field of investigation, comparable in magnitude to the field being presently investigated using adiabatic demagnetization of paramagnetic salts, might be opened.

Purposes of the Present Investigation

The purposes of this work are to demonstrate a brute force nuclear polarization; to determine the angular momentum of the compound nucleus In^{116} in the state corresponding to the 1.46 eV neutron capture resonance; to obtain any cryogenic information as to the temperature of the paramagnetic cooling salt used which may be available; and to develop a part of the requirements for nuclear demagnetization as a step toward the possible eventuality of carrying out such experiments.

¹⁸M. A. Ruderman and C. Kittel, Phys. Rev. 96, 99 (1954).

CHAPTER II

THE "BRUTE FORCE" METHOD, USING NEUTRONS

General Requirements of the Method

A system of nuclei is said to be polarized if the quantity

$$f_N = \frac{\bar{I}_z}{\bar{I}} = \frac{1}{\bar{I}} \frac{\sum_i m_i e^{m_i g_N \beta_N H / kT}}{\sum_i e^{m_i g_N \beta_N H / kT}} \quad (2)$$

differs from zero. We here concern ourselves only with the case of a simple magnetization of a paramagnetic nuclear spin system whose hamiltonian in a magnetic field is given by (1). In (2) above, m_i is the magnetic quantum number for the z component of the nuclear spin, ranging from $-I$ to $+I$, I is the nuclear spin in units $\hbar$, H is the applied magnetic field, which is assumed to be in the positive z direction, k is Boltzmann's constant, and T is the absolute temperature; other quantities have the same symbols as before.

If we consider only this simple magnetization process, the fractional magnetization of a system of nuclear spins may also be written

$$f_N = \frac{2I+1}{2I} \coth \frac{2I+1}{2I} \alpha - \frac{1}{2I} \coth \frac{\alpha}{2I} \quad (3)$$

where $\alpha = \mu H / kT$. This is the well-known Brillouin¹⁹ formula for the

¹⁹L. Brillouin, J. de Phys., 8, 74 (1927).

magnetization of a paramagnetic system of spins and magnetic moments. For the case $\alpha \ll 1$, which is generally applicable in this discussion, (3) reduces to⁵

$$f_N = \frac{1}{3} \frac{I+1}{I} \frac{\mu H}{kT} \quad (4)$$

Limitations

Since μ is of the order of 10^{-23} erg/gauss and k is of the order of 10^{-16} erg/degree, we immediately see that quite large values of H/T are required for substantial nuclear polarization by this method; values of the order of $H/T \sim 10^7$ gauss/degree are clearly required for large polarizations. This would correspond to a field of 100 kilogauss (the highest constant field thus far attained) at a temperature of 0.01°K (not far above the lowest temperatures which have been maintained for reasonably long periods). A serious difficulty immediately becomes apparent; the field must be maintained simultaneously with the temperature at the low value, and since such temperatures can only be reached by the adiabatic demagnetization process at present, it is clear that the nuclear sample must be maintained in the strong field while the cooling agent (coolant salt) is maintained in approximately zero field after the demagnetization. This may be accomplished in two ways; by separation of the nuclear sample and the cooling salt so that the field conditions are fulfilled, or by using a coolant salt which preferably contains the nuclei to be studied and which has a highly anisotropic spectroscopic splitting factor, cooling being obtained by rotating the salt crystal to that direction in which its magneti-

zation is quite small in the magnetic field²⁰. This latter approach is difficult to apply except in very special circumstances²⁰, and the first method (separation) is therefore considered here. It is difficult to conceive of a separation less than ~ 10 cm, and one is thus confronted with the problems of transferring heat to the paramagnetic salt. Among the heats to be transferred are the heat of magnetization given off when the nuclei of the sample drop down to their increased occupation of the lower energy levels, as well as those which arise due to eddy current heating (if the sample is metallic) when the field is applied, and due to the normal influx from the surroundings by various processes. The process of heat conduction has fundamental limitations in that all materials which might be used as heat conductors have thermal conductivities which decrease with decreasing temperature at low temperatures; thus for a given geometry and heat input, there is no gain after the temperature of the coolant becomes small compared to the temperature drop along the conducting material. On the basis of these same considerations, the times involved for the transient heat inputs (magnetization heat, eddy current heat) may also become excessive. On the other hand, the use of large magnetic fields may lead to difficulties with regard to eddy current heating, since in general for the larger magnetic fields, rates of change of field must become larger if a reasonable time for application of the field is not to be exceeded. Thus a rather careful approach to such problems in

²⁰J. W. T. Dabbs and L. D. Roberts, Phys. Rev. 95, 307 (1954).

a given experiment is indicated.

Relaxation Times

Another time enters our considerations; this is the time required for the nuclear population to change from essentially an equal distribution over the magnetic energy levels to the final configuration with larger populations in the lower lying states. Clearly if this nuclear spin-lattice relaxation time τ is too long, it prohibits the performance of an experiment. Although in crystalline, non-conducting, non-magnetic materials τ may be very large^{21,22}, it has been shown²³ that the addition of very small amounts of paramagnetic impurities have a very strong effect in reducing τ . In metals, theoretical^{22,24,25,26} and experimental^{27,28} work has demonstrated that τ values of the order of one second may reasonably be expected near 1°K, with a 1/T temperature dependence. There is also a possibility of a temperature independent contribution to the relaxation processes, which at sufficiently low temperatures would prevent

²¹I. Waller, Zeits. f. Physik 79, 370 (1932).

²²W. Heitler and E. Teller, Proc. Roy. Soc. A 155, 629 (1936).

²³N. Bloembergen, Physica 15, 386 (1949).

²⁴J. Korringa, Physica 16, 601 (1950).

²⁵A. W. Overhauser, Phys. Rev. 89, 689 (1953).

²⁶H. S. Gutowsky, Phys. Rev. 83, 1073 (1951).

²⁷N. Bloembergen, Physica 15, 588 (1949).

²⁸N. J. Poulis, Physica 16, 373 (1950).

further increase in τ as the temperature is lowered^{24,28}. Generally speaking, τ is larger for non-metals, although the situation is by no means untenable.

Thermal Conductivities

Most metals are characterized by a peak in their thermal conductivities at temperatures around 2 - 20°K, followed by a rapid decrease as the temperature is further reduced. However, the very low temperature conductivity is generally proportional to the absolute temperature for metals, as contrasted to thermal conductivities following approximately a T^3 law for many non-metals at very low temperatures. Single crystals of such non-metals as sapphire²⁹ could be used for heat conductors in the region to be considered, because they also exhibit a strong peak in thermal conductivity at roughly 10 - 20°K, but would be very expensive, and at the very low temperatures, would be inferior to most polycrystalline metals. The peak thermal conductivities in both types of materials usually are of the order of 10 watts/cm sec.³⁰ Pure superconducting metals, on the other hand may be quite poor heat conductors at temperatures well below their transition temperatures³¹, and this behavior has led to the suggestion that if a part of the heat conduction link between the nuclear sample and the coolant salt were made of a superconducting metal whose superconductivity

²⁹R. Berman, Proc. Roy. Soc. (London) A 208, 90 (1951)

³⁰NBS Circular 556, (U. S. Government Printing Office, Washington 25, D. C., Sept. 1954).

³¹K. Mendelssohn and C. A. Renton, Phil Mag. 44, 776 (1953).

could be "turned on and off" by application of a field greater than the critical field, one would then have a means of isolation of the nuclear stage of a nuclear demagnetization. It would appear from the above discussion that a metal is a very natural choice for the nuclear sample as well as for the heat conducting link. This gives an additional advantage; there is then no difficulty about the heat contact problem at the nuclear sample, although it may be an important problem at the salt.

Choice of In¹¹⁵

Nuclear Requirements

When polarized slow neutrons are captured or scattered by nuclei of spin $I \neq 0$ whose spins are oriented at random, the cross section is independent of the degree of neutron polarization. If, however, both neutron and nuclear spins are polarized, a change of cross section may occur, reflecting the spin dependence of the neutron-nuclear interaction. Assuming the neutron energy to be near a resonance of the compound nucleus of angular momentum either $J_+ = I + \frac{1}{2}$ or $J_- = I - \frac{1}{2}$ for S neutrons, the polarization dependent cross sections σ_+ or σ_- are given by Eqs. (5a) and (5b) respectively¹⁵.

$$\sigma_+ = \sigma_0 \left(1 + \frac{I}{I+1} f_n f_N \right) \quad (5a)$$

$$J_+ = I + \frac{1}{2}$$

$$\sigma_- = \sigma_0 (1 - f_n f_N)$$

$$J_- = I - \frac{1}{2} \quad (5b)$$

Here f_n is the neutron polarization and f_N is the nuclear polarization defined by Eq. (2). The neutron polarization is similarly defined as $f_n = \bar{S}_z/S$ where S is the neutron spin. The bar in $\bar{S}_z$ refers to the average in the sense of Eq. (2) along the preferred direction, namely, that of the magnetic field.

From the observation of whether the neutron cross section increases or decreases with, e.g., the polarizations of the two spins in question parallel, one is able to infer that the angular momentum of the given resonance is $J_+ = I + \frac{1}{2}$ or $J_- = I - \frac{1}{2}$. For a neutron energy near to more than one resonance, the resultant change in cross section may be due to resonances of different J and one can only infer which J value is predominant in the neutron interaction at the given energy. In the region of neutron energy below 1 eV, and especially near the thermal energy range for nuclear reactors, intense beams of highly polarized neutrons can be produced. Thus, if we restrict ourselves, for the present, to energies near thermal, it is desirable to utilize nuclei with well isolated resonances in or near thermal neutron energies.

The observation of the angular momentum of the compound nucleus at a resonance may be compared with similar information obtainable from studies of cross section as a function of energy. The Breit-Wigner one level formulation³² of the cross section σ_T is, for neutrons,

³²G. Breit and E. Wigner, Phys. Rev. 49, 519 (1936); H. A. Bethe, Rev. Mod. Phys. 9, 69 (1937).

$$\sigma_r = g\pi\chi_r^2 \frac{\Gamma_n \Gamma_r}{(E-E_r)^2 + (\Gamma_n + \Gamma_\gamma)^2/4} \quad (6)$$

where

$$g = 2J+1/(2S+1)(2I+1) \quad (7)$$

and E is the neutron energy, E_r the resonance energy, Γ_n the neutron width, Γ_γ the width for γ -ray emission, and χ is the neutron wave length divided by 2π . The width $\Gamma_r = \Gamma_\gamma$ if (6) is to describe the neutron absorption cross section $\sigma_r = \sigma_a$, and $\Gamma_r = \Gamma_n$ if (6) is to give the neutron scattering cross section $\sigma_r = \sigma_s$. Particle emission (other than neutrons) is not considered here and potential scattering is not taken into account as it is not necessary for this discussion.

When the incoming neutron energy $E = E_r$ (6) reduces to

$$\sigma_r = 4\pi\chi_r^2 \frac{\Gamma_n \Gamma_r}{(\Gamma_n + \Gamma_\gamma)^2} \cdot \frac{2J+1}{(2S+1)(2I+1)} \quad (8)$$

Let us now consider the case $\Gamma_r = \Gamma_n$ and $\Gamma_\gamma \ll \Gamma_n$; i.e., we are investigating a scattering resonance. The neutron width Γ_n cancels out leaving

$$\sigma_s = 4\pi\chi_r^2 \cdot \frac{2J+1}{(2S+1)(2I+1)} \quad (9)$$

Thus if the cross section σ_s is measured at resonance in good resolution, and if I , S and χ_0 are known, we may obtain the angular momentum of the compound nucleus for the given resonance. Although such measurements are very difficult, the discussion does correctly imply that for scattering resonances J may be obtained from phase and cross section

measurements. It is also useful to consider the case of a strongly capturing resonance. With $\Gamma_r = \Gamma_\gamma$ and $\Gamma_\gamma \gg \Gamma_n$, (8) becomes

$$\sigma_a = 4\pi\lambda^2 \frac{2J+1}{(2S+1)(2I+1)} \frac{\Gamma_n}{\Gamma_\gamma} \quad (10)$$

Even if the capture cross section is now measured in good resolution, knowing S , I and λ , one can at best obtain the product $(2J+1) \Gamma_n / \Gamma_\gamma$ and other measurements are then necessary to obtain the width values. For this case the use of polarized particles is especially suitable in providing information about the compound nucleus. It should be mentioned that a technique for measuring σ_s / σ_a for strongly absorbing resonances has been developed by Brockhouse³³, which can be used to obtain J values for such resonances. However, this technique cannot be considered reliable in all instances³⁴; but it is of interest to compare the results of measurements made directly on neutron resonances, as above, with the type of measurement involved in the present work.

It is clear from the above discussion that it is highly desirable to have as the nucleus under study one which is characterized by a substantial thermal absorption cross section and a single, well-isolated capture resonance as near thermal energies as possible. The sample should preferably be a metal from heat transfer and relaxation time requirements,

³³B. N. Brockhouse, Can. J. Phys. 31, 432 (1953).

³⁴B. N. Brockhouse, private communication to L. D. Roberts.

and should have as large a value of μ/I as possible to reduce the difficulty of polarizing the nuclei. If one is to work with thermal neutrons, it is imperative that only one resonance (if not at thermal energies) should contribute appreciably to the neutron cross section at thermal energies.

Comparison of Various Nuclides

A tabulation of certain nuclear properties is given in Table I for some nuclei of interest. The nuclides represented are chosen on the bases that they require no more than 500,000 oersteds/degree K for 1 percent polarization; that they have a non-negligible cross section at thermal energies, and that there are apparently well separated resonances at or near thermal energies. It is virtually certain that several nuclei which possibly fulfill these criteria are absent because of lack of knowledge of their magnetic moments, or because of poor data as to neutron cross sections. The data used in preparing Table I were obtained from the neutron cross-section curves given in AECU-2040³⁵ and the tabulations in the Nuclear Data Tables³⁶.

The choice of In^{115} from these data is supplemented by the following facts; it is a metal, can easily be obtained in high purity, and a soldered heat contact is easily made. The next resonances are well sepa-

³⁵AECU 2040 (Offices of Technical Services, Dept. of Commerce, Washington 25, D. C., 1952).

³⁶Nuclear Data Tables - NBS Circular 499 (U. S. Government Printing Office, Washington 25, D. C., Sept., 1950).

TABLE I

COMPARISON OF NUCLIDES

Nuclide or Element	$\frac{\mu(I+1)}{I}$ (Nuclear Magnetons)	Thermal Neutron Cross-section (barns)	Energy of Neutron Resonance (s) Near Thermal	H/T Required (Oersted/°K) for 1 percent Polarization
H ¹	8.37	35	-	87,500
Li ⁶	1.64	970	2.5×10^5 eV	500,000
F ¹⁹	7.89	3.9	3.3×10^4	103,900
Na ²³	3.70	3.0	2700	221,900
Sc ⁴⁵	6.11	12	prob. neg.	134,200
Mn ⁵⁵	4.86	13	345	168,700
Co ⁵⁹	5.98	41	125	137,100
In ¹¹³	6.71	small in natural In	1.80	122,200
In ¹¹⁵	6.72	198	1.46	122,000
Cs ¹³³	3.31	50	5.9	247,700
Eu ¹⁵¹	4.76	4500	-0.011, 3.3	172,200
Re	4.41	98	2.1	185,900
Tl ²⁰³	4.83	11.6	260	169,800
Bi ²⁰⁹	4.99	6.6	-	164,300

rated, not in energy, but by the fact that the widths, etc. of the higher resonances are such that the cross section at thermal energies is almost entirely due to the 1.46 eV resonance³⁷. A J value for this resonance has been published³⁵; but subsequent high resolution measurements of the cross section as a function of energy³⁸ are not able to confirm this value. Thus a measurement of the type envisioned here, which establishes the J value, is quite desirable. In addition, it should be noted that for his first discussion of nuclear demagnetization, Simon³ chose this metal as an example, even though the neutron properties of the material were of no consequence for his discussion.

There are two disadvantages to this choice; first, indium is a superconductor, and this leads to difficulties with regard to heating as it undergoes this transition; secondly, there is an energetic β^- activity in In^{116} which provides further heating difficulties. These points will be discussed later in detail.

Choice of Cooling Salt

Temperature Requirements

The cooling salt must fulfil several requirements in order to carry out nuclear polarization experiments satisfactorily. The primary requirement is, of course, that it be capable of cooling the In to a sufficiently

³⁷See Eq. (62), Chapter VII.

³⁸H. H. Landon, Private communication.

low temperature that sufficient polarization for the purposes at hand is obtainable, with the available magnetic field. It has been established in previous experiments at Oak Ridge National Laboratory⁸ that it is possible to obtain 10,000 oersted at the nuclear sample with the cooling salt 12.5 cm. away in essentially zero field; for this first experiment in "brute force" polarization, it was deemed desirable to utilize this arrangement again. It is then necessary to consider the minimum polarization which is sufficient for the purposes at hand; it will be shown later that a polarization of 1 to 2 percent is sufficient. Thus from the requirement that H/T must be 122,000 oersted/degree for 1 percent polarization as shown in Table I, we see that a temperature range of 0.04°K to 0.08°K is necessary as a minimum requirement, the lower temperature being, of course, greatly preferred.

Paramagnetic salts suitable for adiabatic demagnetization may be characterized by several parameters. At sufficiently high temperatures, the isothermal magnetic susceptibility usually follows the Curie-Weiss law

$$\chi = \frac{C}{T - \theta} \quad (11)$$

where C is the Curie constant and θ is the Weiss constant which may be either positive or negative. The specific heat of most paramagnetic salts is characterized by a lattice term of the form BT^3 , and in a suitable temperature range, by a term of the form b/T^2 which is a high temperature approximation to a Schottky type specific heat anomaly. Quite often,

suitable salts have two such anomalies, so that it is rather difficult to describe the specific heat without resort to graphs. In Fig. 1 we have given, in comparative form, the results of two groups of workers on Ferric ammonium alum by plotting the entropy against the absolute temperature on a logarithmic scale in the region of interest. The specific heat of the salt is then given by the slope of the curve at the point of interest. Suitable lines delineating the regions attainable in our apparatus have been included. We are forced here to consider only those regions of H/T attainable with the electromagnet used for previous nuclear polarization work^{7,8}. The maximum H/T available for demagnetization has been arbitrarily set at $\sim 16,000$ gauss/degree for the case of pumping the liquid to 0.99°K with an oil diffusion pump, and to $\sim 13,000$ gauss/degree if only the backing pump is used. This limit was set to prevent a repetition of a previous coil failure on the magnet. It is seen that there are large discrepancies between the work of the two groups with regard to the absolute temperature and specific heat; however, the agreement is much better with regard to the magnetic temperature T^* , defined by

$$T^* - \theta = KC/\chi \quad (12)$$

where K is a calibration constant determined at liquid He temperature and χ is the measured magnetic susceptibility of the paramagnetic salt specimen. The discrepancies of Fig. 1 may perhaps be regarded as fairly typical of the difficulties encountered in absolute temperature measurements below 0.1°K . The curves in Fig. 1 are marked as follows: the subscript 0 refers to the Oxford work of Kürti, Simon and Squire, which was

Fig. 1. Entropy—Temperature Diagram— Ferric Ammonium Alum in Zero Field.

carried out at Bellevue in 1937 and reported by Cooke³⁹. The subscript L refers to the work of Steenland, DeKlerk, Potters and Gorter⁴⁰, done at Leiden in 1949. Certainly it may be said that either set of measurements led to results which would predict satisfactory results for our experiments, since we may expect temperatures in the range $0.013 - 0.042^{\circ}\text{K}$ and specific heats between $\sim 0.3R$ and $1.7R$ per mol. degree. It should be pointed out that the curves of Fig. 1 are taken from a published graph giving a 'best curve' in the case of the Leiden results, and determined from measurements made on published curves of S/R vs. T^* and T^* vs. T in the case of the Oxford work. Neither curve may, therefore, be considered to be the best representation possible of these results, but both are quite adequate for our purposes here, namely, to indicate the present knowledge of the salt under consideration. It should be noted that both curves in T have roughly the same general shape below $S/R = \ln 2$, at which point some sort of break in the curves might reasonably be expected on the basis of the energy level configuration for the salt^{39,41} which is thought to be three doublets.

Specific Heat Requirements

We have already mentioned that the specific heat values which may

³⁹A. H. Cooke, Proc. Phys. Soc. (London) A 62, 269 (1949).

⁴⁰Steenland, DeKlerk, Potters and Gorter, Physica 17, 149 (1951).

⁴¹P. H. E. Meijer, Physica 17, 899 (1951).

be derived from the curves of Fig. 1 lie between $\sim 0.3R/\text{mol. deg.}$ and $1.7R/\text{mol. deg.}$. As will be shown later, we may expect the sample size to be ~ 0.01 mol. and the heat input to be of the order of 25 erg/min. On this basis we would expect warm-up rates between $0.0011^\circ/\text{hr}$ and $0.006^\circ/\text{hr}$; these are entirely adequate for present purposes, since the temperature of the salt would not change by more than about 30 percent in the course of an hour at worst, and at best (based on the Oxford results) the temperature change would only be about 3 percent per hour. Thus we conclude that Ferric ammonium alum ($\text{Fe}(\text{NH}_4)(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$) is a suitable salt for the experiments reported here.

Thermal Conductivity of the Salt

Another important consideration for the choice of the paramagnetic salt to be used is its thermal conductivity; however, it would appear that very little is known of the thermal conductivity of salts, except in isolated instances. There is at present no particular reason for supposing that any one salt should be much better than another. However, it is well to note that the thermal conductivity of $\text{K Cr} (\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$ has been measured by Bijl⁴² in a single crystal at liquid He temperatures, and found to vary according to a $T^{2.3}$ power law, and surprisingly enough, to depend by about a factor 5 on the rate of cooling to liquid N_2 temperatures (77°K). The conductivity at 1°K was of the order of 10^{-2} watt/cm deg.

⁴²D. Bijl, *Physica* 14, 684 (1949).

Garrett⁴³ has performed measurements below 1°K on the same material, and found the thermal conductivity to be

$$K = 5.3 \times 10^{-2} T^3 \text{ watt/cm. deg.} \quad (13)$$

This was interpreted to be a lattice conductivity, with a phonon mean free path of ~ 0.5 mm.

Thus we see that at temperatures of $\sim 0.01^\circ\text{K}$ the heat transfer through a 1 cm. cube of such a salt (in the form of a single crystal) is only about 0.1 erg/sec. deg., and that it is extremely difficult to transmit heat at such temperatures. There is some hope that the thermal conductivity of such materials might rise again at the very lowest temperatures⁴⁴ by a process called "exciton conductivity" giving a $1/T^2$ conductivity. Garrett⁴⁵ comments that the theoretical basis for this conclusion is shaky.

The thermal conductivity of powdered samples has been investigated (incidental to other work) by Goodman⁴⁶ using a similar arrangement to that of Mendoza⁴⁷. Goodman found that the thermal conductivity of the salts used could be represented by

⁴³C. G. B. Garrett, *Phil. Mag.* 41, 621 (1950).

⁴⁴A. Akheiser and I. Pomeranchuk, *J. Phys. (USSR)* 8, 216 (1944).

⁴⁵C. G. B. Garrett, *Magnetic Cooling* (Harvard University Press, Cambridge, Mass., 1954), p. 90.

⁴⁶B. B. Goodman, Thesis, (University of Cambridge, 1952) unpublished.

⁴⁷E. Mendoza, *Ceremonies Langevin-Perrin* (Paris, 1948), p. 53.

$$K = 3 \times 10^{-3} T^3 \text{ watt/cm. deg.} \quad (14)$$

with no evidence for a "barrier" at the metal-salt interface in contrast to the earlier experiments⁴⁷.

It has been suggested⁴⁸ that in view of the order of magnitude difference between powders and single crystals, the salt be grown from solution onto the heat conducting elements. This suggestion has been followed for a portion of the work reported here.

⁴⁸E. Mendoza, private communication; Howling, Darnell and Mendoza, Phys. Rev. 93, 1416 (1954).

CHAPTER III

CRYOGENIC DESIGN OF THE EXPERIMENTS

Introduction

In this chapter we shall discuss the detailed arrangements developed for the solution of the problem of obtaining and maintaining the nuclear polarization. To achieve our objective, it is necessary to examine in detail the various sources of heat to which the combined nuclear sample-cooling salt arrangement is subjected in the course of an experiment, and to evaluate the effect of these sources on the final temperatures of the nuclear sample.

Generally, one may state that it is desirable to obtain near-thermal equilibrium between the nuclear sample and the cooling salt since this represents the lowest possible temperature of the nuclear sample. In addition it is necessary that near-thermal equilibrium be attained in a time short compared to the duration of the experiment. These conditions permit the attainment of a nuclear polarization if the nuclear sample is in a strong magnetic field. Maintaining the nuclear polarization requires that the heat capacity of the cooling salt be much greater than the total heat input introduced in the course of an experiment, so that the temperature rise of the cooling salt in this time is sufficiently small.

Our discussion will thus take up the various sources of heat, both transient and steady, then after a description of some general character-

istics of the equipment pertinent to these problems, will proceed to an examination of several sample assemblies, after which a detailed study of the final sample assembly used is made with regard to the problems brought up in the last paragraph above.

Heats to be Absorbed by the Cooling Salt

We list here the heat sources pertinent to the actual performance of a nuclear polarization experiment. The first group of sources appear in the initial phases of the demagnetization of the salt and the magnetization of the nuclear sample. They are

1. Specific heats of the indium metal and other portions of the sample.
2. Eddy currents during demagnetization.
3. Heat associated with transitions into the superconducting region while demagnetizing.
4. Condensation of He exchange gas on salt.
5. Vibration or other heating associated with moving the sample.
6. Cooling associated with transitions out of the superconducting region as the nuclei are magnetized.
7. Eddy current heating during the magnetization of the nuclei.
8. The actual heat of magnetization of the nuclei.

After a quasi-steady state condition has been reached, there remain a number of heat sources in effect which determine how long the

nuclear polarization may be maintained. These are

1. Radiation from the walls at the liquid helium temperature or other temperatures warmer than the sample.
2. Gas conduction from these walls.
3. Conduction through the mounting.
4. Vibration heating.
5. Eddy current heating due to magnetic field ripple.
6. Heating due to the nuclear reaction used to detect the polarization.

Calculations on the effects produced by these heats are not really feasible except for a definite sample design. These calculations will be discussed later.

Previous Development of the Demagnetization Apparatus

A design for a suitable apparatus for indirect cooling of nuclear samples had been developed for a previous experiment on the polarization of Sm^{149} by the Rose-Gorter method at Oak Ridge National Laboratory⁸. The apparatus consisted of an arrangement at the graphite reactor suitable (metal) Dewar flasks, pumping systems, and a large electromagnet for the purpose of performing demagnetizations starting near 1°K . A diagram of this apparatus is shown in Fig. 2. The arrangement was similar to that for a normal adiabatic demagnetization experiment except for a few special features developed for this type of experiment. First, no provision was

Fig. 2. Cryostat for Indirect Cooling - Nuclear Polarization Studies.

made for removing the Weiss electromagnet from the vicinity of the flask or vice versa after the demagnetization; instead, the same magnet was then used to supply the polarizing field for the nuclei. Secondly, provision was made for raising and lowering the sample so that either the cooling salt or the nuclear sample could be centered in the gap of the Weiss magnet. This was accomplished by means of an elevator mechanism for raising and lowering the 1 inch diameter (70 Cu - 30 Ni) sample tube through a sliding "O" ring seal at the top of the cryostat with a rack and pinion which was equipped with a hand crank and accurate stops at the two ends of the travel of 5 inches. This is shown in its mid-position in Fig. 2. A magnetic shield surrounding the lower end of the Dewar flasks reduced the field on the cooling salt when the nuclear polarizing field was applied, thus allowing the samples to be kept cold.

The upper end of the sample tube was attached to valve "A" of Fig. 2 by means of an "O" ring joint, so that the valve could be removed, allowing insertion of the sample assembly and radiation shields. The horizontal outlet could be attached to either of two pumping tubes ("U" of "L" in Fig. 2) by means of another "O" ring seal, the unused tube being stopped off by a blanking plate. A second right angle valve (B) completed the connection to a fast oil diffusion pump (MCF 300) which was used to remove the He exchange gas. The intervening space between valves A and B was let up to atmospheric pressure whenever the sample tube was raised or lowered.

The magnetic shield was constructed of Armco iron, and consisted

of two hollow cylinders whose inner and outer radii formed a geometric progression. The upper end of the outer shell was folded inward to the inside diameter of the inner shell in such a way as to preserve the spacing between the shells and to present a surface nearly parallel to, and about 1 inch away from the lowest surface of the pole tips of the electromagnet. The foldover arrangement is due to a suggestion of Mr. R. H. Ward and is thought to be important to the effectiveness of the shield, although no tests were made on this point. It was found that the shielding was very complete up to about 11,000 oersted in the 1.9 inch gap of the electromagnet, and deteriorated substantially in higher fields, giving 360 oersted in the shield for a field of 15,400 oersted in the gap.

The Heat Link as Previously Used

In the Sm^{149} experiment, the dual sample was made as follows⁴⁹: The nuclear sample (a paramagnetic salt) was compressed inside a thick (~ 1 mm) walled Cu box which was silver soldered to a Cu tube of $1/4$ inch outside diameter and $\sim 1/2$ mm wall thickness. The lower end of the tube was silver soldered to an Ag cup which was essentially two hollow concentric cylinders with one end closed into which the coolant salt was compressed. This sample assembly was suspended by a group of nylon threads under spring tension in the sample tube.

Although no definitive studies have been made, experimental evidence

⁴⁹Reference 8, figure 1.

seems to indicate that heating is induced by vibration of adiabatically demagnetized samples⁵⁰, and this has in general been borne out in a large amount of work in our laboratory. It should be mentioned at this point than an error appeared in the publication⁸ on nuclear polarization in Sm^{149} with regard to heat influx. A figure of 20-40 erg/min was quoted; unfortunately, this referred to another type of sample. In the sample assembly described⁴⁹ the heat leak was actually of the order of 300 erg/min, and was probably somewhat higher while the nuclear sample was being bombarded by the neutron beam; this accounts for the discrepancy between the temperature of the nuclear sample and the cooling salt which was obtained from the cross section change observed in the Sm^{149} experiment.

Developments for Present Experiments

In an attempt to improve the situation, a new sample arrangement has been developed and used in most of the experiments reported here. A number of significant improvements have been obtained. These improvements were in the areas of reduction of vibration heating and eddy current heating, and in improving the quality of the thermal contact to the cooling salt.

Reduction of Vibration Heating

The general approach here was to make a more rigid mounting for

⁵⁰Darby, Hatton, Rollin, Seymour, and Silsbee, Proc. Phys. Soc. (London) A 64, 861 (1951).

the nuclear sample-cooling salt assembly so that this unit was less likely to move with respect to its immediate surroundings. This was accomplished by enclosing the unit within a rigid metal cage and mounting it on rather heavy plastic (heat insulating) supports. This cage was in turn suspended by a zig-zag arrangement of nylon threads under spring tension.

It has been found previously⁵¹ that a "guard" salt could be of great value in reducing heat input to a demagnetized sample. This idea was used here because of the rather heavy plastic supports for the inner sample assembly; at temperatures of the order of 0.8°K , it was clearly demonstrated in heat leak experiments that the conductivity of these supports was quite substantial. The thermal conductivity of such materials might be expected to follow a T^3 dependence, and thus become very effective heat insulators provided the entire insulator is cold; if one end were relatively hot, the strong temperature dependence soon concentrates almost the entire temperature drop at the colder end, and the insulator is no longer effective. The cage, or guard salt (Ferric ammonium alum) was effective in performing the function of cooling the "warm" ends of the solid plastic supports to a sufficiently low temperature ($0.1 - 0.2^{\circ}\text{K}$) to prevent undesirably large amounts of heat conduction.

Eddy Current Heating

One early set of experiments (Experiments I, Chapter VI) used a

⁵¹A. H. Cooke and R. A. Hull, Proc. Roy. Soc. (London) A 181, 83 (1942).

simple modification of the Sm^{149} sample described above, with a 0.060" thick strip of In metal (folded so as to present three thicknesses to the neutron beam) directly replacing the Cu box of the Sm sample. In the final assembly described below, a very great improvement in eddy current heating was obtained by a geometrical redesign from this viewpoint.

Heat Contact to the Salt

The sample assembly previously described used a silver cup into which the salt was compressed in a hydraulic press to 150 or more atmospheres. This gave moderate heat transfer rates only. In the final inner sample assembly, a technique of growing the salt sample onto wires soldered to the nuclear sample was used. This has a number of advantages: (1) The heat contact at the salt-metal interface surely must be much more intimate, since it was observed that there was a very slight etching of the Ag surface by the iron alum solution. This was so slight that probably no important changes in the salt composition or its magnetic properties occurred, though no experiments were done to check this point. (2) The salt formed round the wire in quite large crystallites, and was almost completely clear optically. This implies that the thermal conductivity of the salt should approach the conductivity of a single crystal, which is much higher than that for a pressed powder. (3) Each wire is very likely well insulated from its neighbors, although some inadvertant electrical contacts were probably present between some of the wires. (4) The wires were flattened, and by this means a metal surface area of $\sim 40 \text{ cm}^2$

was introduced into the salt in a distributed manner, since the wires were distributed rather uniformly throughout the body of the salt. Both sides of each flattened wire were in contact with the salt. In contrast, the cups previously used contained almost twice as much salt and had a surface area of about 25 cm^2 in contact with the powdered salt, the salt density being 92.5 percent of the crystalline density in a typical sample. (5) The salt sample could easily be made spherical in contrast to the case where pressing dies had to be constructed and used. This is of some advantage in the measurement of magnetic susceptibilities.

The Final Sample Assembly

The inner sample assembly finally developed is shown in Fig. 3. Mechanically, the assembly was based on a tube of fluorothene plastic, which has a very small cross section for neutrons. The lower end of this tube was serrated internally and the iron alum salt grown in such a way as to rigidly attach itself to the plastic tube. No difficulties due to different expansion coefficients have appeared in a large number of demagnetizations. At the upper end, the tube was slotted to receive the bundle of 20 In plates 0.010 inch thick. The bundle of plates was insulated by a zigzag arrangement of very thin fluorothene foil between the plates. This was found to give excellent electrical insulation. An Ag wire 0.018 inch in diameter was flattened to a width of $\sim 1.5 \text{ mm}$ over a length of $\sim 2 \text{ mm}$ and this tab soft soldered to each of the In plates.

Fig. 3
SAMPLE ASSEMBLY

(The plates were of normal isotopic abundance⁵²; i.e. 95.84 percent In¹¹⁵, 4.16 percent In¹¹³. It is known that both these isotopes are radioactive with half-lives of the order of 10^{14} yrs and can thus be treated as stable nuclei for all practical purposes here.) The 20 wires were individually coated with baked araldite over their central portions to prevent electrical contacts. The Ag wires were also flattened at their lower ends over a length of 6 cm to a width of ~ 1.7 mm. To insure that no reduction in the wire cross section occurred in the flattening, this was done by pressing between flat steel plates rather than by rolling. The 20 wires thus had a total surface area in the flattened lower ends of about 40 cm^2 . These flattened ends were curled up into spirals in as uniform a manner as possible with all of the twenty spirals placed, in random orientations, in the volume which would later contain the salt sphere. The salt was then grown from aqueous solution around this assembly of flattened wires, the outside of the salt being formed by a greased hemispherical ended cup of plastic which was slipped off the salt after it had crystallized. The excess salt at the top was ground off, leaving a fairly accurate sphere. The surface was lightly coated with Apiezon "N" stopcock grease to prevent dehydration of the salt.

The guard cage and its associated salt surrounded the inner sample assembly and is also shown in Fig. 3. An indication of the spring loading on the mountings is shown.

⁵²R. F. Hibbs, AECU-556; reference 36.

With the above background it is possible to turn now to detailed calculations with respect to the various heat inputs and their effects.

Transient Heat Inputs

Specific Heat of the Indium, Silver Wires, etc.

The specific heat of metals at very low temperatures is usually quite small and proportional to the absolute temperature. For indium⁵³, the atomic heat is given by $C = 3.2 \times 10^{-4} T$ cal/mol.deg and for silver⁵⁴, by $C = 1.5 \times 10^{-4} T$ cal/deg.mol. We have neglected the T^3 contribution of the lattice and consider only the electronic specific heat. If we assume that the demagnetization is carried out so slowly that equilibrium exists between salt and metal, the entropy increase of the salt due to these specific heats on cooling to, say, 0.03°K from 1°K is given by

$$S = (n_1\gamma_1 + n_2\gamma_2) T_1 - T_2 \quad (15)$$

where n is the number of moles of metal, γ is the coefficient of T in the atomic heat and the subscripts 1 and 2 refer to the two metals. Similarly, the total heat absorbed by the salt is given by

$$Q = \frac{n_1\gamma_1 + n_2\gamma_2}{2} T_1^2 - T_2^2 \quad (16)$$

For our sample, these calculations give $Q = 2.7 \times 10^{-5}$ cal = 1130 erg.

⁵³J. R. Clement and E. H. Quinell, NBS Circular 519, pp. 89-97 (Oct. 1952)

⁵⁴W. H. Keesom and J. A. Kok, Physica 1, 770 (1934).

The entropy change is 2260 erg/deg which is the equivalent of $\sim 2 \times 10^{-3}$ R/deg.mole (of cooling salt). The specific heat of the other materials in the assembly may be neglected.

Eddy Current Heating During Demagnetization

The eddy current heating during this phase of the experiment is calculated in a straightforward manner, neglecting the fields produced by the eddy currents compared to the applied field. We shall consider for the moment only the heat generated in the Ag spirals within the salt sample. If the dimensions of one of the flattened wires are L, W, and D where $L \gg W > D$, and the length L is assumed to be curved into a spiral within the salt, it may be shown, by an approximate average over the assumed random distribution of angles of the planes of the spirals, that the heating is given for slow, linear field changes by

$$Q \approx \frac{\dot{B}^2 10^{-9}}{3 \rho \pi} \left[\frac{D^3 WL}{2} + \frac{W^3 DL}{2} \right] t \text{ ergs}$$

$$\approx \frac{\dot{B}^2 10^{-9}}{6 \rho \pi} W^3 D L t \text{ ergs} \quad (17)$$

where $\dot{B}$ is the rate of change of the field in oersted/sec., t is the time duration of the change, and ρ is the resistivity of the metal in ohm-cm, all dimensions being in cm. For our sample and a demagnetization involving a 15,000 oersted change in $7\frac{1}{2}$ min, and using an approximate value for the resistivity of the Ag, viz, 2×10^{-8} ohm-cm, independent of the temperature, we obtain a heating of only 7 ergs; therefore, we may neglect heating in

the connecting wires as well as within the salt.

In the flat indium plates, a similar calculation to that above may be made in which no average over angular position is required; thus the equivalent of Eq. (17), if $L \gg W \gtrsim D$, is

$$Q = \frac{B^2 10^{-9}}{6 \rho} D^3 W L (\cos^2 \theta + R \sin^2 \theta) t \text{ ergs} \quad (18)$$

where the symbols and units are the same as before, θ being the angle between the dimension W (the intermediate dimension) and the field direction, and $R = W^2/D^2$. Using the above, for purposes of calculation we assume the field varies linearly from 3000 oersted to 270 oersted in 6.8 min, at which time the In plates become superconducting, and do not give further eddy current heating. The resulting heating is found to be 9 ergs with ρ taken to be $6.4 \times 10^{-9} \text{ ohm cm}^{55}$, independent of the temperature, and θ assumed to be 4° . This heating is, therefore, also negligible if we take 7.5 min to perform the demagnetization.

Heat Associated with Superconducting Transition During Demagnetization

This heating is to be expected because of the entropy difference between the normal and the superconducting states, the latter having an entropy which is lower. The difference is obtained from the fact that the Gibbs free energy difference is just that given by the field energy difference because of the total expulsion of flux from the body of the

⁵⁵S. Valentiner, Z. Physik 115, 11 (1940) and Meissner, Franz, and Westerhoff, Ann. Physik 13, 505 (1932), Table III.

superconductor, viz,

$$G_n - G_s = \frac{H_c^2}{8\pi} \cdot V \quad (19)$$

where subscripts s and n refer to the superconducting and normal states respectively, V is the volume of the body, and H_c is the critical field required to destroy the superconductivity. The entropy difference is then derived from the relation $S = -\partial G/\partial T$ thus giving⁵⁶

$$S_n - S_s = -\frac{V H_c}{4\pi} \frac{\partial H_c}{\partial T} \quad (20)$$

Therefore the heat involved in the transition is, ideally,

$$Q = -VT \frac{H_c}{4\pi} \frac{\partial H_c}{\partial T} \quad (21)$$

If we assume that H_c is given by

$$H_c = H_0 (1 - (T/T_c)^2) \quad (22)$$

we obtain

$$Q = \frac{V H_c H_0}{2\pi} \left(\frac{T}{T_c} \right)^2 \quad (23)$$

At low temperatures ($T \ll T_c$) we may replace H_c by H_0 with sufficient accuracy so that for indium ($H_0 = 269.2 \text{ } \phi$, $T_c = 3.368^\circ$)⁵⁷ we obtain

⁵⁶D. Shoenberg, Superconductivity (Cambridge University Press, 1952) pp. 56-59.

⁵⁷A. D. Misener, Proc. Roy. Soc. (London) A 174, 262 (1940).

$$Q = \frac{(269.2)^2}{2\pi} \cdot \frac{T^2}{(3.368)^2} V \quad (24)$$

and for our sample volume of 5.16 cc. we have a heating in the transition being considered of //

$$Q \approx 5249 T^2 \text{ erg}$$

The transition would be expected to occur at the temperature at which the stray field of the magnet at the In sample reached the critical field; in the initial stages of a demagnetization the salt temperature is almost proportional to the field, and since the stray field of the magnet at the In position was about 1/6.8 of the field in the gap, one might expect the transition to occur at a temperature given by

$$T = 6.8 T_1 \frac{H_c}{H_1} \quad (25)$$

For typical experimental conditions this is $0.11^\circ - 0.15^\circ \text{K}$. Thus the heating involved here is at most about 120 ergs, and the salt entropy change, under equilibrium conditions, is thus $\sim 10^{-3} \text{ R/deg.mol.}$

Condensation of He Gas Onto Salt

It is quite difficult to assess the amount of heat given up to the salt in this process. It may be assumed that initially there is a gas pressure of the order of 10^{-6} mm Hg which condenses onto the sample, and that the heat of condensation is roughly the heat of vaporization of bulk liquid He, which is $\sim 20 \text{ cal/mol}$. A volume of 100 cc contains $\sim 1.7 \times 10^{-9} \text{ mol He}$ at 1°K and 10^{-6} mm Hg . This gives rise to a heating of only 1.4 erg.

However, a dynamic condition probably exists in which gas is desorbed from the walls and adsorbed onto the sample.

Heating Associated with Lowering the Sample Tube

After demagnetization is complete, the sample assembly is lowered a distance of 12.5 cm to bring the nuclear sample into the magnet gap. Careful tests established that when the sample tube was lowered slowly (~ 1 inch/min.) and evenly, no detectable temperature rise of the sample was produced, even though the He bath level was raised and warmed somewhat by contact with the warmer portions of the sample tube and Dewar walls. It was found that the ionization gauge connected to the upper end of the closed-off sample tube registered a decrease in pressure during this operation. This observation does not necessarily imply, however, that the He gas pressure at the lower end of the sample tube decreased, since a measurement of the partial pressure of He would be required to infer this. If such an inference could be drawn, one might suppose that this was due to adsorption onto the additional tube wall which is submerged in the liquid He during the lowering.

Cooling Due to the Superconducting Transition

When the reverse process to the transition into the superconducting state is performed, a cooling is obtained whose magnitude is given by Eq. (24) above. Since the transition in which we are interested occurs at temperatures below 0.05°K , this effect is quite small. However, it has been shown that an eddy current heating may occur in thin plates parallel

to the field whose magnitude is given by⁵⁸

$$Q = \frac{H^2 - H_c^2}{8\pi} V \quad (26)$$

when the transition is induced by suddenly raising the field to a value $H > H_c$, then allowing the transition to occur. Here, "suddenly" means rapidly compared to the time required for the critical field to penetrate the sample. For purposes of estimation we use this result of Lifshitz⁵⁸, together with the approximate rate of change of field during this process. We obtain an eddy current heating of ~ 3500 erg in this manner, for $H = 1.1 H_c$. This was a very approximate estimate, but serves to show the importance of this effect. Because of the importance of this effect, great care was taken to prevent this heating by going through the transition in steps of ~ 0.3 oersted.

Eddy Current Heating During Nuclear Magnetization

This heating of the In plates, which occurs as the field is further increased above the value required to quench the In superconductivity, is readily calculated from Eq. (18) modified to take into account the fact that batteries were used and field changes were made with a rheostat so that the field is approximately given by

$$B = B_i + (B_{i+1} - B_i)(1 - e^{-t/\tau}) \quad (27)$$

This calculation gives a heating for the i^{th} step

⁵⁸I. M. Lifshitz, J. Exp. Theor. Phys. U.S.S.R. 20, 834 (1950) quoted by T. E. Faber, Proc. Roy. Soc. A 219, 75 (1953), p. 86.

$$Q = \frac{B_{i+1}^2 - B_i^2}{12 \rho \tau_i} D^3 W L 10^{-9} (\cos^2 \theta + R \sin^2 \theta) \text{ ergs.} \quad (28)$$

Here B_{i+1} and B_i are the field limits and τ_i is the time constant of the magnet circuit for the i^{th} step. By recasting this equation in terms of the voltage applied, inductance and resistance of the magnet windings, and resistance in the rheostat, one obtains

$$Q = \frac{C^2 E^2 D^3 W L 10^{-9}}{12 \rho L} (\cos^2 \theta + R \sin^2 \theta) \sum_{i=1}^N \frac{1}{r-i}^3 \quad (29)$$

where the magnet circuit resistance is $\alpha - \beta_1 = \beta(r - 1)$, L is the magnet inductance, E is the applied voltage, C is the (assumed) ratio of field to current for the magnet, and N is the number of steps used in bringing the rheostat resistance to zero. An approximate calculation over the range 300 oersted to 10,000 oersted under our conditions yields a heating of 260 ergs, which corresponds to an entropy rise for the salt under equilibrium conditions of $\sim 10^{-2}$ R/deg.mol. In the above we have neglected possible magneto-resistance effects which would tend to reduce this heating somewhat.

Heat of Magnetization of the Nuclei

Corresponding to the nuclear magnetization given by Eq. (3) the entropy is given by

$$S/R = \ln \frac{\sinh \frac{2I+1}{2I} \alpha}{\sinh \frac{\alpha}{2I}} - \alpha f_N \quad (30)$$

For small polarizations, the first term reduces to $\ln(2I+1)$ and the change in entropy is given by the second. Hence from the second law we may write the heat involved as

$$Q = T \Delta S = \frac{3I}{I+1} f_N^2 T R/\text{mol} \quad (31)$$

for small polarizations where T is the absolute temperature at which the polarization is carried out. For indium, $I = 9/2$, and for our sample of 0.1429 mol (16.40 gm), we obtain a heating of 573 ergs for a polarization of 2.1 percent carried out at a temperature of 0.043°K , which corresponds to a change in the salt entropy under equilibrium conditions of 1.8×10^{-2} R/deg.mol.

Steady Heat Inputs

Radiation

The radiation heat from warm walls, etc. may be calculated from the Stefan-Boltzmann formula. For a sample at such a low temperature that back radiation may be neglected we may approximate the result by

$$\dot{Q} = \gamma A T^4$$

where γ is the Stefan-Boltzmann constant, A the surface area of the sample, and we have assumed the emissivity to be unity, and that the sample

is completely surrounded by walls at the temperature $T \gg T_0$, the sample temperature. For our sample a surface area of $\sim 30 \text{ cm}^2$ is appropriate, with a wall temperature of at most 1.3°K . Thus this heat input amounts to 0.3 erg/min , a very modest quantity.

Gas Conduction from the Walls

This has already been mentioned. An estimate of a few ergs/min, based on experience, is accepted here.

Conduction Through the Mounting

Little is known of the thermal conductivity of plastics in the temperature range below 1°K . However, a crude estimate may be made from a value for Perspex by Berman⁵⁹. He gives a value of $\sim 5 \times 10^{-4} \text{ watt/cm}^\circ\text{K}$ at $\sim 2.5^\circ\text{K}$ and states that the variation at the lowest temperatures seems to be linear in T at such temperatures. This would then give conductivities of $\sim 2 \times 10^{-5} \text{ watt/cm}^\circ\text{K}$ at temperatures of $\sim 0.1^\circ\text{K}$, the average temperature of our guard salt. The heat input to our sample by this means would be approximately 180 erg/min on this basis, if the sample were at 0.05°K . However, there is some experimental evidence, using samples similar to our sample, that this heat influx is only 80 erg/min with a 0.015° temperature difference at 0.8°K . Projecting this to the low temperature, we calculate a heat current of 33 erg/min , which appears to be in much better agreement with the general run of experiments on samples of

⁵⁹R. Berman, Proc. Roy. Soc. (London) A 208, 90 (1951).

type.

Vibration Heating

So little is known at present about this source of heating that calculations are pointless. However, it may be mentioned that the inner sample assembly mounting was designed with rigidity in mind, and though the heat input to the guard salt was usually several hundred erg/min, the heat input to the inner sample was always much lower. This indicates, in view of the calculation above, that possibly no vibration heating occurred to the inner sample assembly, although a substantial amount may have been involved in the warm-up of the guard salt.

Eddy Current Heating Due to Magnetic Field Ripple

This was reduced by using a large capacitor (11,000 μ fd) across the magnet while operating with the motor generator set, and by utilizing batteries during those portions of the experiments where the nuclei were being polarized.

Heating Due to Neutron Capture

It was found experimentally that a very substantial heating occurred when the direct neutron beam from the nuclear reactor was allowed to fall on the nuclear sample. This amounted to ~ 225 erg/min for a $9/32$ inch diameter beam, or ~ 1350 erg/min for a $3/8$ " x 1" beam. This is presumed to be due to the 13 second isomer In^{116} which emits a 2.95 Mev β^- in addition to the capture γ -rays. Neglecting the latter and using a value

of the cross-section for formation of this isomer of 52 barns, we calculate a flux of 5×10^6 neutrons/cm² sec, after making a suitable allowance for the energy carried off by the neutrinos in the reaction. This estimate probably should be reduced somewhat because the heating from the γ -ray flux in the neutron beam makes a contribution. It was observed that there was a buildup in the heating over a period of an hour; this is attributed to the 54 min isomer which has a thermal cross section of about 145 barns, and has β^- radiation of ~ 0.85 Mev associated with its decay. The flux value determined in this way is entirely reasonable, though certainly not accurate. It should be noted that the data of Cooke³⁹ were used to calculate all the heat leaks mentioned above. The means which were taken to reduce this type of heating will be discussed in Chapter IV, and results obtained under various conditions described in Chapter VI. It need only be stated that this heat input can be reduced to a very small value by a suitable arrangement of the neutron apparatus.

Effects of Heat Input

Steady-State Effects

We are now in a position to assess the effects of the many sources of heat input just discussed. It is clear that we may neglect many of them, although others are quite important. For the purposes of these calculations we need only consider the effects of the steady heat inputs, since the transient heat input effects associated with the inner assembly

are fairly small, and are complete in times which are short compared to the duration of an experiment (~ 2 hours). Of course, we use the transient heating calculations to estimate the entropy change of the salt during the preparations for the neutron portion of the experiments. Thus we have in the main to consider the "background" heating which is probably of the order of 40 erg/min, and, in all likelihood, is mostly due to conduction through the plastic mounts. We are interested in the temperature difference between the In sample and the cooling salt under the conditions of the experiments carried out. We, therefore, make the pessimistic assumption that all heat reaching the salt arrives via the In plates and the Ag wires, and whenever an assumption is made to simplify the calculations, we shall use that assumption which maximizes the temperature difference between the two ends of the inner sample assembly. Thus the calculations should lead to an upper limit to the temperature drop occurring between the nuclear sample and the cooling salt.

The basic heat conduction equation is quite often written⁶⁰

$$\rho C \frac{\partial T}{\partial t} = K \nabla^2 T \quad (32)$$

where ρC and K are the specific heat (per c.c.), and thermal conductivities respectively and are independent of the temperature. In the case where $\rho C = f(T)$ and $K = g(T)$, the equation may be written

⁶⁰H. S. Carslaw, Mathematical Theory of the Conduction of Heat in Solids, (Dover, New York, 1945).

$$\frac{\partial}{\partial t} \int_0^T f(T) dT = \nabla^2 \int_0^T g(T) dT \quad (33)$$

and in the particular case where $\rho C = \alpha T^{\nu}$ and $K = \beta T^n$ ($\nu \neq -1$; $n \neq -1$) it becomes

$$\frac{\alpha}{\nu+1} \frac{\partial T^{\nu+1}}{\partial t} = \frac{\beta}{n+1} \nabla^2 (T^{n+1}) \quad (34)$$

In the situation at hand, all thermal conductivities and specific heats are strongly temperature dependent. We shall take these dependences into account where it is sufficiently simple to do so, and use approximations in line with the criterion mentioned above otherwise.

Heat flow within the salt. In this case the thermal conductivity of the salt goes as T^3 and the specific heat is in the neighborhood of a maximum (refer to Fig. 1 assuming that the warm-up starts near an initial entropy corresponding to 16,000 gauss/degree). We assume that ρC is independent of temperature, since this is nearly true, and see immediately that if the T^3 dependence of K is taken into account, the equation to be solved is nonlinear. We, therefore, assume K to be independent of the temperature and use the lowest value of K which might be encountered.

Since the flattened wires are more or less uniformly distributed throughout the salt, we may assume each one of these wires is straight and surrounded by its portion of the total salt volume. In addition, it is simpler to assume that the rectangular wire is replaced by a circular wire of radius r_0 such that it has the same surface area as the

flattened wire, the volume of the cylinder of salt surrounding it being adjusted slightly so that the thickness of the salt layer is the same as before. The specific heat of the salt layer is also adjusted by the same factor in the opposite direction to compensate for this. For this particular calculation, the wire is assumed to have a very large thermal conductivity compared to the salt so that no variation along the axis exists. We also assume that the heat input is divided uniformly among the 20 wires. Under these conditions and with suitable boundary conditions, at the inner and outer radii of the salt layer, the solution of Eq. (32) for one of the cylindrical salt-wire arrangements is of the form

$$T = \sum_i e^{-\lambda \alpha_i^2 t} [a_i J_0(\alpha_i r) + b_i N_0(\alpha_i r) + A \ln r + B + D r^2 + \gamma t] \quad (35)$$

where $\lambda = K/\rho C$, and the α_i are the roots of

$$\begin{aligned} Z'_0 &= -\alpha_i Z_1(\alpha_i r_0) = 0 \text{ and} \\ -\alpha_i Z_1(\alpha_i r_1) &= 0 \end{aligned} \quad (36)$$

r_0 and r_1 being the inner and outer radii of the salt layer and

$Z_p(x) = a J_p(x) + b N_p(x)$, the Bessel function notation of Janke-Emde⁶¹ being followed. The constants A, D and γ are listed below:

⁶¹E. Janke and F. Emde, Tables of Functions, 3rd. Ed., (B. G. Teubner, Leipzig and Berlin, 1938).

$$A = - \frac{\dot{Q} r^2}{2KV_s} \quad D = \frac{\dot{Q}}{4KV_s} \quad \gamma = \frac{\dot{Q}}{\rho CV_s} \quad (37)$$

In the above V_s is the volume of the salt, i.e. $\pi(r_1^2 - r_0^2) \ell_s$ and $\dot{Q}$ is the heat influx via a single wire. The constant B is evaluated by assuming the initial temperature distribution to be the same as in the large time solution. Let T_0 be the temperature on the outer surface ($r = r_1$) the assumed salt cylinder at time zero. Then

$$B = T_0 + \frac{\dot{Q} r_1^2}{2KV_s} \ln r_1 - \frac{\dot{Q}}{4KV_s} r_1^2 \quad (38)$$

and the large time solution for the salt temperature at $r = r_0$ (i.e. at the wire surface) is

$$T_s = T_0 + \frac{\dot{Q} r_1^2}{2KV_s} \ln (r_1/r_0) - \frac{\dot{Q}}{4KV_s} (r_1^2 - r_0^2) + \gamma t \quad (39)$$

or more simply

$$T_s = T_0 + \Delta T + \gamma t \quad (40)$$

The connecting Ag wires. Here both the thermal conductivity and specific heat are proportional to the absolute temperature. We can use, therefore, Eq. (34) with $\nu = n = 1$. The problem is one dimensional, and since the specific heat is very small for metals at low temperatures, any transients will die away in very short times. We, therefore, set the specific heat of the wire equal to zero. Since the temperature of the salt can be expected to change extremely slowly, this assumption is justified; we may thus obtain quite a close approximation to the correct

temperature drop. If the heat input to the warm end of the wire is $\dot{Q}$, and the cross sectional area is a , the equation to be satisfied when this boundary condition is taken into account is

$$\frac{\partial T^2}{\partial x} = \frac{2\dot{Q}}{\alpha_{Ag} a} \quad (41)$$

and straightforward integration gives $T^2 = 2\dot{Q}x/\alpha_{Ag} a + f^2(t)$, where $f(t)$ is the temperature of the heat sink (i.e., the salt) and is given above as $(T_0 + \Delta T + \gamma t)$. Thus we find a temperature at the warm end of the wire ($x = \ell$) to be

$$T = \sqrt{2\dot{Q}\ell/\alpha_{Ag} a + (T_0 + \Delta T + \gamma t)^2} \equiv T_W \quad (42)$$

Conduction within the In plates. The equation to be satisfied here is the same as for the Ag connecting wires, except in two dimensions and with the addition of a source term on the right representing the generation of heat within the In by the nuclear reaction. The remainder of the heat, which is conducted by the plastic mounts and by gas, may also be considered as being introduced uniformly into the plates. If we call the heat introduced per unit area into the plates $\dot{q}$ and let $\gamma = \alpha_{In} d$ be the thermal conductivity taking into account the thickness d of the plates, and make the further assumption that the plates are circular segments to eliminate angular variation, the geometry is essentially cylindrical and Eq. (34), again neglecting the specific heat, may be written

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T^2}{\partial r} \right) = - \frac{2\dot{q}}{\gamma} \quad (43)$$

This equation has the solution

$$T^2 = - \frac{\dot{q} r^2}{2 \gamma} + A \ln r + g^2(t) \quad (44)$$

If we take into account the fact that no heat flows in through the outer edge we may evaluate A to be $\dot{q} R^2 / 2 \gamma$ where R is the radius of the outer edge. The third term may be evaluated from the fact that $T = T_w$ (Eq. (42)) at the inner radius r_2 where the connection to the Ag wire is made. We thus obtain for the temperature at the outer radius R of the plates

$$T = \left[(T_0 + \Delta T + \gamma t)^2 + 2 \dot{q} l / \alpha_{Ag} a + \frac{\dot{q} R^2}{2 \gamma A_p} \ln \left(\frac{R}{r_2} \right) - \frac{\dot{q}}{2 \gamma_{In} A_p} (R^2 - r_2^2) \right]^{\frac{1}{2}} \quad (45)$$

where the symbols have the meanings given previously, $\dot{q}$ being the total $\int \dot{q} dA$ received per second by each indium plate and A_p being the area of one of the plates.

We may now calculate the indium temperature from Eq. (45) under certain assumed conditions. We use the data on temperatures and specific heats of the salt presented in Fig. 1 for 16,000 gauss/degree corrected for an entropy increase of 0.02 R/mol due to transient heat inputs, first taking the Oxford³⁹ and then the Leiden⁴⁰ data, together with the dimensions etc., of our sample assembly, and use an assumed heat influx $\dot{q} = 0.033$ erg/sec (40 erg/min for the 20 plates). We also assume a thermal conductivity for indium of 0.6 T watt/cm deg, which is 3.5 times smaller than that of Mendelssohn and Rosenberg⁶² to allow for thermal magneto-

⁶²K. Mendelssohn and H. M. Rosenberg, Proc. Phys. Soc. (London) A 65, 388 (1952).

resistance effects⁶³. An estimated thermal conductivity of $0.15 \text{ T watt/cm deg}$ for silver, based on data of White⁶⁴, is also used. We thus obtain

$$T_{\text{In}} (\text{Oxford data}) = \left[(0.0374 + \Delta T + 3.5 \times 10^{-7} t)^2 + (0.0188)^2 + (0.009)^2 \right]^{\frac{1}{2}} \quad (46)$$

The first term in the brackets is associated with the salt, ($\Delta T \sim 10^{-5} \text{ }^\circ\text{K}$), the second with the wires, and the third with the In plates. The physical interpretation of the quantities in the parentheses of the second and third terms is that these are the temperature drops which would appear if the "cold" end of the part in question were at the absolute zero of temperature.

If we evaluate this expression at $t = 5000 \text{ sec}$, we obtain

$$T_{\text{In}} (\text{Oxford data}) = 0.043^\circ\text{K} \quad (47)$$

essentially independent of the time during an experiment. Similarly, using the Leiden data, the result is

$$T_{\text{In}} (\text{Leiden data}) = \left[(0.0158 + 0.00021 + 1.96 \times 10^{-6} t)^2 + (0.0188)^2 + (0.009)^2 \right]^{\frac{1}{2}}$$

which, evaluated at 2000 and 5000 seconds, gives

$$T_{\text{In}} (\text{Leiden data}) = \begin{cases} 0.0290^\circ\text{K} (2000 \text{ sec}) \\ 0.0334^\circ\text{K} (5000 \text{ sec}) \end{cases} \quad (49)$$

⁶³K. Mendelssohn and H. M. Rosenberg, Proc. Roy. Soc. (London) A 218, 190 (1953).

⁶⁴G. K. White, Proc. Phys. Soc. (London) A 66, 844 (1953).

Transient Effects

In order to assess the thermal time constant of the system, we note that the nuclear spin-lattice relaxation time, and the thermal relaxation times within the In plates and the Ag wires are all quite short, the latter two being small because of the small specific heats. We, therefore, need consider only the time for thermal diffusion within the salt. This may be developed from the first line of Eq. (35). We consider only the first term of the sum, since this will give the longest time constant, and we are not interested in any particular initial temperature distribution. Equations (36) which are obtained from two of the boundary conditions on that part of the temperature represented by the first line of Eq. (35), are simultaneously satisfied if the relation

$$T_1(x_n) = T_1(kx_n) \quad (50)$$

is satisfied where

$$T_p(x) \equiv J_p(x)/N_p(x) \quad (51)$$

again following the notation of Janke-Emde⁶¹ where $k = r_1/r_0$ and $x_n = \alpha_n r_0$.

The value of $x_1 = 3.52$ is found from the graph given there (p. 207) and

the value $k = 1.9$ for the salt cylinders. Therefore $\alpha_1 = 66.4 \text{ cm}^{-1}$

($r_0 = 0.053 \text{ cm}$) and the diffusion of heat has the time dependence

$$e^{-x\alpha_1^2 t} = e^{-4409Kt/\rho c} \quad (52)$$

If we insert the Garrett value (Eq. (13)) for the thermal conductivity

at T_0 as given by the Oxford group, along with the specific heat derived

from their data, or the equivalent values using the Leiden data, all of which may be obtained from Fig. 1, we find a relaxation time of 5 seconds using the Oxford data and 17 seconds using the Leiden data. Thus we see that the calculated relaxation time is quite short compared to the time required for an experiment.

General Conclusions

The calculations given above indicate that one might reasonably expect the nuclear sample to reach temperatures in the region of the desired values mentioned previously in times short compared to the duration of an experiment, and to remain at these temperatures for periods of several hours. In this sense the design should be expected to be a suitable one.

We have seen that the most important transient sources of heat may be associated with items 4, 6, 7, and 8 in the list on page 32, and that an assumed entropy increase of 0.05 R/mol.deg for the salt, due to these sources, is probably correct within a factor 2. Under steady state heat flow conditions, the most important sources of heat are probably the plastic mounting conduction, gas conduction, and the nuclear reaction, in that order. Our estimate of 40 erg/min for the total heat input is based to a large extent on the first mentioned source.

In the analysis above, the effects of eddy current heating, etc., on the guard cage were not taken into account; as will be seen in Chapter

VII, it is possible that in the present instance, these heatings may have been quite important insofar as their effect on the temperature of the upper portion of the cage and the consequent heat flow through the plastic mountings was concerned. However, since in future experiments a redesign of the guard cage would surely be made, it was not considered necessary or desirable to carry out a detailed analysis of this portion of the sample assembly here.

CHAPTER IV

NUCLEAR DESIGN OF THE EXPERIMENTS

Thickness of the In Sample

The sample of In metal may be considered as a sample in which cross section changes are to be determined by a neutron transmission measurement. Since the transmission is given by

$$\mathcal{V} = e^{-Nt \sigma_0 (1 \pm f_I f_N f_N)} \quad (53)$$

where N is the number of nuclei per cm^3 , t is the thickness, σ_0 is the cross section corresponding to no polarization, and f_I is the spin factor depending on whether J is $I + \frac{1}{2}$ or $I - \frac{1}{2}$, we see that the fractional change in transmitted intensity is given, for counting rates $\bar{C}$, by

$$|\Delta \mathcal{V}| = \frac{|\Delta C|}{\bar{C}} = 2 \sinh Nt \sigma_0 |f_I f_N f_N| \quad (54)$$

for a reversal of the relative spin orientations. Thus for small polarizations, the change to be observed is directly proportional to the product $Nt \sigma_0$, the number of factors e of neutron attenuation in the nuclear sample, as well as to the product of the two polarizations $f_I f_N$. It is, therefore, to our advantage to make the nuclear sample as thick as possible consistent with intensity requirements, since we may thus reduce the nuclear polarization required for a given size effect.

However, it is also clear that by the very nature of the experi-

ment, an unlimited time is not available to use for the performance of the measurements; therefore, a more careful consideration is indicated. Rose and Shapiro⁶⁵ have shown that there is, for a given background, an optimum thickness in a fixed area beam, which minimizes the counting time required for a given accuracy in the determination of the transmission of a sample. The same type of analysis may be applied to the case of a change in the transmission of a sample, such as concerns us here. If we let the quantity T of Rose and Shapiro⁶⁵ represent the transmission ratio for the change of cross section represented by the factor $1 + f_I f_n f_N \equiv 1 + \gamma$ and define $y = Nt \sigma_0$ and $x = \gamma y$ to correspond to their notation, we obtain instead of their Eq. (22a) the following expression whose root is the optimum value of y :

$$m e^{2\gamma y} \left[1 + \frac{1}{2\gamma} - \frac{1}{\gamma y} \right] \left[\frac{1}{f_1} + \frac{1}{f_2} \right] + \frac{e^{\gamma y}}{2f_1} \left[1 + \frac{1}{\gamma} - \frac{2}{\gamma y} \right] + \left[\frac{m+1}{f} + \frac{m}{f} \right] \left[\frac{1}{2\gamma} - \frac{1}{\gamma y} \right] = 0 \quad (55)$$

Here m is the relative background and the f_1 are the f_1 of Rose and Shapiro (their Eq. (18)) multiplied by the factor e^y . Inspection of the above equation shows very quickly that for reasonable, i.e., small, values of γ , the optimum value of y is very close to 2, regardless of the value of m , the result being determined almost solely by the brackets containing γ and y .

⁶⁵ M. E. Rose and M. M. Shapiro, Phys. Rev. 74, 1853 (1948).

In order to enhance the size of the effect to be observed, it was decided to make the sample of In so that $Nt\sigma_0$ was $\gtrsim 2$ for the neutron energy used. This, of course, increased the counting time required for a given accuracy of determination. For a relative background $m = 0.1$ and $y = 2.75$, Eqs. (19) and (21) of Rose and Shapiro⁶⁵ may be used to show that, if the relative background were the same in the two cases, the total counting time must be extended only 13 percent over the optimum time to overcome the decrease accuracy incurred by the reduction of the counting rates. The optimum distribution of the counting times is essentially the same in both cases, being ~ 0.41 , 0.41 , and 0.18 for the two determinations and the background respectively. The above figures, on the basis of comparing the transmission of 90 percent polarized and depolarized neutrons through a sample polarized to the extent of 1.6 percent include the factor $I/I+1 = 9/11$, giving $\gamma = 0.012$. If the spin of the neutrons is reversed, γ becomes 0.024 , and the total counting time for 10 percent accuracy in the determination of γ becomes, for an incident flux of 30,000 neutrons/min, ~ 80 minutes instead of ~ 320 minutes for the "one-sided" effect. Thus it is very desirable to reverse the polarization of the neutrons, if possible. This point will be discussed later in detail.

Neutron Polarization Methods

There are at present only two well-developed methods for polarizing neutrons, and both of these are restricted to the thermal region. In

addition, very large neutron fluxes are only available in the thermal region from nuclear reactors. Thus we shall consider only these two methods at present, and shall confine ourselves to thermal neutrons.

Transmission Through Magnetized Fe

The question of polarization by this method has been discussed by Stanford et al⁶⁶ and in references cited there. Stanford et al found for example, that a Maxwellian distribution of neutrons at 495°K from the Oak Ridge graphite reactor showed an average polarization of 32 percent when passed through 4 cm of Fe magnetized to essential saturation when impinging on a $1/v$ detector. This work was carried out in connection with a previous experiment on polarized nuclei⁷. The advantage of the method lies in the intensity available, since 4 cm of Fe has a transmission of about e^{-4} . However, the rather small polarization and lack of energy selection makes any experiment using this method an intrinsically more complicated one, since averages over energy must be used. In addition, departures from the $1/v$ law in the sample being studied may reduce the average effective polarization substantially. Nevertheless, the method was used to obtain a small effect in In¹¹⁵, as will be described later (Experiments III, Chapter VI).

Diffraction from Magnetized Fe₃O₄

This method was also studied by Stanford et al⁶⁶; they found ex-

⁶⁶Stanford, Stephenson, Cochran, and Bernstein, Phys. Rev. 94, 374 (1954).

perimentally that certain large natural Fe_3O_4 crystals gave polarizations as great as 90 percent in first order reflections from the 220 planes; the second order contribution was rather large and was polarized to the extent of 10 percent in the opposite direction. The polarization obtained is independent of energy except that the ratio of first to second order intensity varies due to the Maxwellian energy distribution incident from the graphite reactor. In the best of the experiments reported here, the difficulty with second-order reflections was largely obviated by a second reflection from the 111 planes of Cu. The reflected intensity is about e^{-6} of the incident intensity. We have used one of the same Fe_3O_4 crystals which was used by Stanford et al⁶⁶ in all work using this method.

Neutron "Flipping" Methods

A means for reversing the predominant direction of the polarized neutron spins relative to the magnetic field is highly desirable, since it doubles the size effect to be observed, and also reduces the total counting time required to obtain a given accuracy by a factor 4, as mentioned above.

Radiofrequency Nuclear Magnetic Resonance

This method is very elegant, and quite suitable for studies of neutron polarization⁶⁷; in practice, however, it is not really suitable

⁶⁷C. P. Stanford, Thesis, University of Tennessee, 1954 (unpublished).

as a method for reliably reversing the neutron spins. The frequency stability requirement on the oscillator (which may have a power output of the order of 1 KW) is of the order of 2 parts per million; this is, of course, easily capable of attainment, at the expense of considerable care. The requirement on the value of the magnetic field in which the oscillator coil is located is the same, namely, ~ 2 parts per million, and to obtain such a field reproducibly is extremely difficult. Therefore, one would have to resort to continuous monitoring of the neutron polarization during all experiments in order to guarantee that flipping is in fact accomplished with efficiencies of 90 percent or greater. The method is for this reason rejected here.

Abrupt Field Reversal

A method which is quite successful for all reasonably slow neutrons was first discussed by Majorana⁶⁸ in connection with atomic beam experiments. He considers a particle of spin $\hbar/4\pi$ passing through a magnetic field such that the three components of field acting on the particle are zero, A (a constant) and Gt , where t is the time. Under these circumstances, he shows that the transition probability for a non-adiabatic process of spin-reversal with respect to the field is given by

$$W(1/2, -1/2) = W(-1/2, 1/2) = e^{-r^2 \mu A^2 / \hbar G} \quad (56)$$

⁶⁸E. Majorana, Nuovo Cimento 9, 43 (1932). English translation available as ADI Document 2229, Armour Research Foundation, Chicago, Ill. (1945).

where μ is the absolute value of the magnetic moment of the particle and h is Planck's constant. This expression was obtained by considering in the asymptotic expressions for the wave function at positively and negatively infinite times; however, Motz and Rose⁶⁹ have obtained the exact solution in a more complicated case, and given the same result. The result is, of course, independent of the relative directions of the neutron path and the field components.

For our purposes we recast Eq. (56) in terms of the fields and field gradients for neutrons of velocity v cm/sec and obtain

$$W(1/2, -1/2) = W(-1/2, 1/2) = e^{-\eta H_1^2/v \nabla_p H_2} \quad (57)$$

In this case η has the magnitude 1.44×10^5 gauss⁻¹ sec⁻¹. The subscripts 1 and 2 refer to any two perpendicular field components, and $\nabla_p H_2$ means the gradient (in gauss/cm) of component 2 along the direction of the path. For 0.075 eV neutrons $\eta/v = 0.038$ gauss⁻¹ cm⁻¹, and we see that for a gradient of 1 gauss/cm, A must be ≤ 0.51 gauss for a transition probability ≥ 99 percent, and must be ≥ 11 gauss for a transition probability ≤ 1 percent. The conditions for "flipping" the spins with respect to the field, or "transmitting" the spin unchanged with respect to the field, are thus established.

This method has been used many times in atomic beam work, and has

⁶⁹L. Motz and M. E. Rose, Phys. Rev. 50, 348 (1936).

been applied to neutrons by Stanford et al⁶⁶ and others⁷⁰. In all previous neutron work, as in some of the work here, various arrangements of iron plates, tubes, etc., and permanent magnets have been used to obtain fields of the nature required for a "Majorana flip". These have been very successful in some cases and not too successful in others. In any case, this approach requires a good bit of empirical adjustment, which is time consuming, in order to change the arrangement back and forth between "flip" and "transmit" conditions. It was fortunately possible, in one of the experiments (Experiment IV), to make use of stray fields together with a system of air-core coils, so that completely reproducible and predetermined field arrangements could be obtained quickly. In this way a changeover from "flip" to "transmit" conditions was possible in less than 30 seconds.

A difficulty presents itself in that it is not possible to get perfectly uniform flipping when the neutron beam is of finite cross sectional area if the field gradients are produced in free space. This follows from the fact that $\text{curl } H = 0$ in free space, and if one component (other than the component along the beam) has a gradient, other components must inevitably also vary as one moves away from the point where H_z of Eq. (57) equals zero. This means that flipping conditions are different in different parts of the beam. In earlier work, it is likely that too-large gradients have often been used, with consequent imperfection in the flipping.

⁷⁰See, for example, D. J. Hughes, Pile Neutron Physics, (Addison-Wesley, Cambridge, Mass., 1953).

Suggestion of a New Method

It should be possible, in view of the difficulty mentioned above, to produce a "perfect" flipping arrangement for large area beams by performing the flipping within a region where $\text{curl } H \neq 0$, i.e., a region carrying current. In practice, this could be accomplished in two ways: (1) by passing the neutron beam through a thin foil of a diamagnetic metal carrying a uniform current perpendicular to the propagation vector of the neutrons; and (2) by passing the neutrons through the equivalent of a foil made of closely spaced, small diameter fine wires. The metal of such a foil must have a low neutron cross section. The foil or wire system should, of course, have large transverse dimensions compared to the transverse dimensions of the beam; this has the further advantage that a uniform field in the beam region would be maintained independent of the distance from the foil out to distances of the order of the smaller of these two dimensions. The method would have a number of advantages; (1) conditions would be essentially uniform over the beam area; (2) large gradients would be possible using thin foils and moderate currents; (3) the system would be tolerant of rather large stray field components, e.g., a stray field of ~ 5 gauss would be permissible with ~ 0.1 eV neutrons for a gradient within the foil of ~ 100 gauss/cm; and (4) very slow neutrons would be "flipped" more easily. The only other requirements to be satisfied are that the field component having the gradient pass through zero within the foil, and that the thickness of the foil be very large compared to the wave length of the neutrons. Then, within the foil, it would be possible to have al-

most exactly the Majorana conditions. It is thought that this method has not been suggested previously. The foil method has not been tried here, because of lack of time, and the fact that a rather suitable flipping arrangement had already been developed.

Neutron Depolarization

It is desirable, for comparison reasons, to depolarize a polarized neutron beam in the course of the experiments. The equivalent of this depolarization might, of course, be obtained by merely removing the magnetic field from the neutron polarizer, or in destroying the nuclear polarization, either by removing the magnetic field on the nuclear sample or by warming the sample to a "high" temperature. None of these techniques is really suitable. If one removes the neutron polarizer field, one introduces the so called "single-transmission" effect, with consequent intensity changes. If the magnetic field on the nuclear sample is removed and reapplied, unnecessary eddy current heating is induced, and the entropy of the coolant salt is increased. The process of recooling the sample after a warm-up requires at least one hour, and exact reproducibility is difficult to obtain. Thus the method of depolarizing neutrons which is used is highly desirable, in that it requires only the introduction, between polarizer and analyzer, of a thin sheet of unmagnetized iron ("shim") which is quite effective⁶⁶. The small attenuation of the beam thus introduced is compensated by placing the same shim just ahead of the counter

during periods when the effects of polarized neutrons are desired.

The technique discussed above has been dealt with in the paper of Stanford et al⁶⁶ mentioned earlier, and was discussed in detail by Stanford⁶⁷.

Neutron Heating in Various Arrangements

The amount of heating caused by the neutron capture in the sample varies widely with the arrangement used in the experiments, because the number of neutrons actually incident on the sample, even for the same final counting rate, depends strongly on this arrangement.

Direct Beam - Fe_3O_4

This arrangement, used in the Sm^{149} experiment mentioned earlier⁸, and shown in Fig. 4, consists in having the direct beam of neutrons from the reactor pass through the nuclear sample, which acts as a neutron polarizer. The beam is then analyzed by diffraction from the 220 planes of a magnetized Fe_3O_4 crystal. Early attempts at observing an effect in In^{115} used this arrangement. It has the advantage of allowing an effective polarization of the neutrons of ~ 90 percent for the first order⁶⁶. However, the second order neutrons are little polarized and relatively intense. This is not a sufficiently strong objection to eliminate the method, since for some samples the strong neutron flux through the sample does not lead to a large heating; however, in In^{115} , strong β^- activities are induced which lead to pronounced heating. In experiments of this type, heating

76

Fig. 4. Neutron Arrangement for Experiments I and II.

at the rate of ~ 1350 erg/min was observed, and could only be reduced by reducing the beam size and counting rate.

Transmitted Beam - Fe

This arrangement consists of a straight beam in which a thick (say 4 cm) block of magnetized Fe serves as the neutron polarizer, the analyzer being the nuclear sample itself, and is shown in Fig. 5. The attenuation introduced by the iron block is about one factor e per cm of thickness. Thus the typical 4 cm block used reduced the heating effect by about a factor 50, giving ~ 30 erg/min for In , which is, for the present experiment, almost satisfactory. This arrangement was used in an earlier experiment⁷ on Mn^{55} . However, the Fe block does not give a definite energy for the neutrons, nor is the average polarization large. This latter fact makes it very difficult to obtain a visible effect, as will be seen in the discussion of this type of experiment on In^{115} in Chapter VI, although a definite effect was found. The method does have the advantage that a rather large counting rate is attainable, so that a relatively small effect may be observed in fairly good statistics. It suffers from the possibility that, in attempting to reverse the neutron spins, one may introduce errors because of possible changes in the single transmission effect when reversing the field applied to the Fe block.

Diffracted Beam Fe_3O_4

A new arrangement, unused previously in the type work described here, has been introduced for the last of the present experiments. It

Fig. 5. Neutron Arrangement for Experiments III.

consists in reversing the order of events along the neutron flight path from that described above under "Direct Beam - Fe_3O_4 ". Here the sample is placed in the diffracted beam from a single crystal of Fe_3O_4 , which acts as the neutron polarizer in this instance. This immediately obviates all difficulty with heating due to the nuclear reaction, since the intensity of (monoenergetic) neutrons striking the sample is reduced by a factor of the order of 400 over the direct beam intensity. Thus a neutron heating of only about 4 erg/min is introduced. The advantage of large neutron polarization is retained, and the counting rate is essentially the same as in the direct beam - Fe_3O_4 case.

Double Crystal Spectrometer-Polarizer

A further improvement of the above arrangement was introduced in those experiments (Experiments IV) where a diffracted beam incident on the indium was used. It was desired to reduce the rather large second order content of the diffracted beam, and also to obviate the necessity of moving the very bulky cryogenic equipment. Both these ends were accomplished by performing a second diffraction from the 111 planes of a single crystal of Cu. The use of the Cu 111 planes was suggested by E. O. Wollan. The second diffraction automatically reduces the second order content of the beam by a factor 4 because of the factor λ^2 in the usual equation⁷¹

⁷¹Halpern, Hamermesh, and Johnson, Phys. Rev. 59, 981 (1941), Eq. (2.11).

for the reflected intensity, the crystal structure factor being the same for the 111 and 222 planes of Cu. Since the 111 spacing for Cu is smaller than the 220 spacing in Fe_3O_4 , the angle for reflection of a given energy is larger. This allows the diffracted beam to be brought back across the original line of the main beam. Thus the nuclear sample was used in the same position as in other experiments, the main beam being absorbed in shielding of appropriate character. For the neutron energy used the amount of second order flux content was calculated to be reduced to about 1.9 per cent of the first order flux by this procedure. The arrangement is shown in Fig. 6.

An important difficulty arose in connection with this arrangement. It was found to be rather difficult to transmit or flip a polarized beam when using the process of the two reflections and passage into the sample position. This difficulty was not fundamental, but arose from the fact that the beam traveled a path which was not symmetrically disposed with respect to the stray field of the low temperature magnet. A similar trouble arose from the asymmetry of the coil placement in the Fe_3O_4 magnetizing magnet. These difficulties were obviated by elimination of all Fe in the region between the two magnets, and modifying the stray fields which existed normally along the neutron path (and which by chance were peculiarly suitable for the purpose), with a set of rectangular Helmholtz-type coils. A graph showing the three components of the stray field is shown in Fig. 7, for a magnet current in the "forward" direction in the Fe_3O_4 magnet. The coordinate system is that shown in the lower left of Fig. 6. It is easily

81

Fig. 6. Neutron Arrangement for Experiments IV.

Fig. 7. Stray Field Components along Neutron Path; Coordinate System as in Fig. 6.

seen that by adding constant fields of ~ 7 and 5 gauss, respectively, to the x and y components, a field zero may be obtained where the z component is zero. The measurements were made near the center of the beam with a Rawson rotating coil fluxmeter, and the coils, which are shown in the photograph of Fig. 8, were designed so that for fields of ~ 7 gauss, uniformity within ~ 0.3 gauss over the area of the beam was obtained except near the ends, where strong fields were already present. Predetermined current settings for the best flipping were obtained by measurements of the double transmission effect using Fe blocks in the low temperature magnet as analyzers for the polarization.

The z components due to both the low temperature magnet and the Fe_3O_4 magnet were ~ 17 gauss near the Cu crystal, and tended to cancel each other when the Fe_3O_4 magnet current was in the "forward" direction. By changing this current to the "reverse" direction, the y (vertical) component, which was due entirely to this magnet was reversed, and the z component was, of course, ~ 34 gauss near the crystal. Under these conditions, the neutron spins were "transmitted" unchanged with respect to the field, which rotated slowly (and approximately uniformly) from the vertical to the horizontal direction as one traversed the neutron path. The double transmission effect was found to be independent of the currents in the coils when the current directions were such as to increase the magnitude of the field along the path. This was taken as evidence that no "flipping" occurred under these conditions. In the nuclear polarization experiments, these coil currents were maintained at a standard value in these directions

84

PHOTO 14149

84

Fig. 8. Neutron Flipping Apparatus.

corresponding to about 6 gauss.

We, therefore, were able to change from one neutron polarization direction with respect to the nuclear polarizing field to the other in ~ 30 sec. by a) reversing the Fe_3O_4 magnet current, and b) manipulating a reversing switch and 2 rheostats controlling the air core coils. In the experiments using this apparatus, these changes were the only ones made to obtain reversal of the relative spin orientation of the neutrons and the In nuclei.

CHAPTER V

AUXILIARY AND ASSOCIATED EQUIPMENT

Cryogenic Equipment

Dewar Flasks

The two concentric helium and nitrogen Dewars were of conventional metal construction, with two exceptions. They could be disassembled at "O"-ring joints, and there was a "displacement foil" of brass in the neutron path to remove the boiling nitrogen in the outer Dewar from the neutron beam. The outer Dewar was continuously pumped by a VMF-20 oil diffusion pump, and the inner (He) Dewar had an arrangement whereby H_2 gas could be admitted to the vacuum space for precooling to liquid N_2 temperature. This H_2 was pumped to ~ 0.05 mm Hg after the precooling and closed off, and the remainder of the evacuation was produced by freezing out at liquid He temperatures.

Helium Pumps

The liquid He bath used as a starting point in the demagnetizations was pumped by a 5 horsepower mechanical pump (Kinney) with a capacity of 40 liters/sec. For an evaporation rate of 200 cc of liquid He per hour, this pump was capable of reducing the vapor pressure to 0.74 mm Hg, which is equivalent to 1.224°K, neglecting pressure drop along the pumping line. In addition, there was installed a large metal booster pump (KB 150 oil

pump) which had a rated capacity of 150 liters/sec with backing pressures ~ 1 mm. This pump produced a bath pressure of 0.11 mm. Hg, which is equivalent to 0.991°K . It was necessary to monitor the voltage across the 4 KW heater in this pump against line variations to insure stable operation.

Pressure Measurements

These were made with three instruments; a mercury manometer, dibutyl phthalate manometer, and a standard thermocouple gauge. The vacuum leg of the two manometers was continuously pumped, and the back pressure corrected for by readings of a thermocouple gauge. The density of the dibutyl phthalate was measured, and simultaneous measurements on the two manometers also used in the calibration of this low pressure manometer. The thermocouple gauge was used for He pressure readings only for relative values to assist in maintaining steady conditions from the KB 150 diffusion pump.

Sample Elevator Mechanism

The sample tube was made of 70 Cu - 30 Ni, and was 1 inch O.D. with a 0.020 inch wall. The wall thickness and material were chosen for low heat conduction. At the upper end of the sample tube an "O" ring sliding joint permitted axial motion so as to raise or lower the sample tube 5". A rack and pinion arrangement permitted very smooth operation between accurately positioned stops. The upper end of the sample tube was attached to an "O" ring joint to a 1 inch Veeco right angle valve. The outlet of this valve could be connected by means of "O" ring joints to either of

two branches of a pumping tube which led to a $1\frac{1}{2}$ inch Veeco right angle valve mounted at the inlet to the MCF-300 oil diffusion pump which was used to remove the He exchange gas just prior to the demagnetization. The demagnetizations were carried out in the upper position, the lower branch of the pumping tube being blanked off. Next, after closing the two valves, the intervening space was let up to atmosphere and the sample tube lowered. It was found that further pumping in the lower position had no effect; therefore, this was not done.

Radiation Shields

To protect the sample from infra-red radiation down the tube, a series of four radiation shields of the ring and disk type were inserted in the tube above the sample assembly. These are shown in Fig. 2. This arrangement was demonstrated to have an attenuation greater than 10^8 for the room temperature radiation.

Susceptibility Measurements

The technique used was the usual ballistic galvanometer and compensated mutual inductance with the salt sample as a core⁷². Because of spatial limitations in Dewars not specifically designed for this experiment, it was necessary to place the two samples of $\text{Fe}(\text{NH}_4)(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$ very close together, and one was faced with the problem of obtaining the

⁷²H. B. G. Casimir, Magnetism and Very Low Temperatures, (Cambridge University Press, 1940) p. 16.

susceptibilities of the two salts individually. This was partially accomplished by making the "main" primary and secondary coils for each salt quite short ($\frac{1}{2}$ inch) and placing the "bucking" mutual inductance at a reasonably large distance from both salt samples. In addition the two "main" sets of coils were separated by a larger distance than the center to center distance between the two salt samples. The residual correction for the effect of one salt at the other's coil was obtained by calibrations at liquid He temperatures with one salt removed, and with both salts present. Thus solutions of a pair of simultaneous equations gave the desired results, while qualitative results could be obtained directly from the galvanometer readings. The galvanometer was a Leeds and Northrup type 2285 e. Each primary coil had 243 turns of number 36 Cu wire, and each secondary coil had 1069 turns of number 40 Cu. The primary current was usually ~ 100 mA. It was recognized that the results of these measurements could not be accepted as other than an indication of the salt susceptibilities, because of the inhomogeneous field of the primary coils over the salt volumes. In addition, the cage salt was not spherical, so that demagnetizing factors could not be accurately taken into account. However, these measurements served the purpose of giving a good indication of the salt susceptibilities, and were quite consistent with expectations. They also indicated that, under a given set of conditions, the warm-up of the samples was quite reproducible.

Weiss Magnet

The low temperature magnet was a conventional oil cooled Weiss magnet with pole pieces shaped according to a prescription of Dwight and Abt⁷³. The magnetic shield, described previously, was mounted below the pole tips. The supply for this magnet was a motor generator set supplying 105 volts at 330 amperes for 16,400 gauss, or for the nuclear polarization itself, a bank of eight 12 volt storage batteries supplying 24 volts at 90 amperes, to give 11,150 gauss. The motor generator set was excited with a simple storage battery and series rheostat arrangement which proved to be quite stable.

Nuclear Equipment

Fe₃O₄ Crystal and Magnet

The Fe₃O₄ crystal used as polarizer or analyzer for the neutrons in some of the experiments reported was one of the same crystals used by Stanford et al⁶⁶. A magnetizing magnet, requiring about 500 watts to produce a field of ~ 2500 gauss on the crystal, normal to the scattering plane, was used. The crystal was 2 inches square and 1/8 inch thick, and reflection was always from the 220 planes. The spacing of these planes is 5.922 Å;⁷⁴ thus a Bragg angle of 10°10' gives a neutron energy in first order of 0.075 e.V. The magnet was mounted on a special spectrometer

⁷³H. B. Dwight and C. F. Abt, R.S.I. 7, 144 (1936).

⁷⁴R. W. G. Wyckoff, Crystal Structures, (Interscience Publishers, New York, 1948).

table made for neutron diffraction experiments. The counter arm was removed for the last of the experiments reported here.

Fe Blocks and Magnet

In some of the work reported here, the heterogeneous beam of neutrons emerging from the ORNL graphite reactor was polarized by transmission through thick (4 cm) blocks of magnetized 1018 cold rolled steel. These steel blocks were oriented so that the magnetic field (11,000 gauss) was applied parallel to the rolling direction. Similar blocks of smaller thickness were used as analyzers to check the polarization of the Fe_3O_4 crystal and the effectiveness of the spin reversal arrangement. This was accomplished by placing these blocks (of 1 and 2 cm effective thickness) in the gap of the low temperature magnet and measuring the shim effect under the two conditions of spin orientation. It was found that under the conditions of the last experiments the flipping efficiency of the arrangement was ~ 91 percent. The polarization cross section for Fe at the wave length ($1.044 \text{ \AA}$) corresponding to 0.075 eV was found to be 0.95 - 1.0 barn in fairly good agreement with previous work (cf. Stanford⁶⁷, Fig. 3).

Boron Filter

In order to determine the amount of second order reflection from the Fe_3O_4 crystal corresponding to a first order energy of 0.075 eV, a laboratory standard BF_3 absorption cell was used. The transmission of this cell is well known, since it has been measured in a number of ways.

The diffracted beam from the Fe_3O_4 crystal, not reflected from the Cu crystal was used. The amount of second order (flux) was found to be 7 percent of the amount of first order after making a small correction for third order content. This figure does not include a correction for the relative efficiencies of the counter at the two energies; however, this is the correct value for use in calculations regarding final results. From this, one calculates that after the Cu reflection, the beam contained effectively 1.7 percent second order, which is small enough to be neglected for most purposes. It should be noted that if one takes the counter sensitivity and the form factors⁶⁷ of Fe_3O_4 into account, the amount of second order found agrees rather well with the amount calculated on the basis of a 495°K ⁶⁶ Maxwellian distribution joined to a $1/E$ slowing down distribution at 0.17 eV ⁷⁰. Dr. E. C. Smith very kindly remeasured the transmission of the BF_3 cell (after these measurements) on the ORNL time of flight neutron spectrometer and found very good agreement with the standard values. The time-of-flight spectrometer was, of course, completely free of second order or other higher energy neutrons, and thus affords a reliable test.

Neutron Counter

The neutron counter was filled with ~ 95 percent B^{10}F_3 to a pressure of 40 cm Hg. Its active region was 2 inches in diameter and 18 inches long, and the counter was surrounded by a heavy shield of paraffin, boron and cadmium. The counter plateau extended from 2250 to 2500 volts; this

voltage was furnished by a regulated power supply. A standard A-1 amplifier⁷⁵ and a cathode follower mounted on the end of the counter were used. The pulse height discriminator was adjusted to transmit virtually all neutron pulses and essentially no gamma ray pulses or noise. A scale of 64 circuit (Atomic Instruments) completed the main counting arrangement. An electric clock, actuated by the counting switch was used to time all counts.

⁷⁵W. H. Jordan and P. R. Bell, R.S.I. 18, 703 (1947).

CHAPTER VI

DESCRIPTION OF EXPERIMENTS

Experiments I

These experiments were performed in November 1953 and March 1954. Some 10 actual attempts were made to observe the desired results.

Sample

The sample used was the simple dumbbell sample described earlier in which the In metal was in the form of a folded strip soldered to the Cu heat conducting tube which led to the Ag salt cup. Ferric ammonium alum was the coolant, and the assembly was simply suspended on nylon strings.

Neutron Arrangement

The direct beam - Fe_3O_4 arrangement, described in Chapter IV and shown in Fig. 4, was used. Thus the experiments were essentially duplications of the Sm^{149} work⁸, with the substitution of the In metal for the Sm salt. These experiments always appeared to give a small effect (change in transmitted neutron intensity with changes in polarization of the neutrons) but this was not outside the statistical errors of the experiment.

Conclusions

It is clear that several difficulties contributed to the failure to observe an effect on the transmitted neutron intensity when the neutrons

were depolarized. These were the large eddy current heating associated with this sample, the substantial heat leak to the sample by any of several mechanisms, and the heating of the In sample by β^- activity induced by the neutron bombardment. The large heatings led to rather short warm-up times which further reduced the opportunities for demonstrating that the effect had been present.

Experiments II

These experiments were performed in July 1954.

Sample

The sample used in these and all succeeding experiments was the sample described and analyzed in Chapter III and shown in Fig. 3.

Neutron Arrangement

The neutron arrangement was the same as in experiments I. However, the first experiment immediately showed the strong effect of neutron induced heating on the warm-up rate of the sample (as determined from susceptibility measurements on the coolant salt). A recalculation showed that an error had been made in a previous calculation on this heating. An attempt was made to reduce the heating by collimating the beam in such a way that the neutron intensity was reduced by a factor of 4. Under these conditions, an effect of 2.8 ± 1.7 percent was observed in one run. The direction of the effect was such that it indicated $J = I + \frac{1}{2}$ as the angu-

lar momentum of the level of the compound nucleus corresponding to the 1.458 eV resonance.

Conclusions

This result was not considered sufficiently reliable, and the neutron heating was considered sufficiently severe to demand a rearrangement of the neutron setup because 1) the counting rate with the reduced beam was small, and 2) the sample warm-up time was shortened to the point that many repetitions of the experiment would have been required to determine the size of the effect with any accuracy.

Experiments III

Neutron Arrangement

In order to reduce the heating caused by the β decay in the nuclear sample, it was decided to sacrifice the advantage of large neutron polarization, at least temporarily, to determine whether or not a reasonable amount of nuclear polarization had been achieved. The neutron arrangement used for these experiments was that shown in Fig. 5, and was described as the transmitted beam - Fe arrangement in Chapter IV. The neutrons were polarized by transmission through a 4 cm thick block of 1018 cold-rolled steel. The rolling direction was along the applied field of 11,000 gauss, which was supplied by a 5 KW Weiss magnet. The parallel plates between the polarizer magnet and the low temperature magnet maintained a field direction transverse to the beam direction. This field was adjusted by

manipulation of a number of permanent magnets bridged across the gap between the parallel plates. It was possible in this arrangement to compare polarized and depolarized beam counts by moving a depolarizing "shim" from just in front of the counter to a position between the two magnets. The neutrons could also be brought to the nuclear sample with their spins "flipped" with respect to the magnetic field by means of a rapid reversal of the field direction (Majorana flipping) in the region controlled by the parallel plate arrangement. It was not possible, however, to change from "transmitted" to "flipped" spins either quickly or reliably in this arrangement, since it was difficult to rearrange the permanent magnets reproducibly. It was also found that the intensity transmitted by the polarizer block was not reproducible on reversal of the field on the block. This was probably due to a fault in the (electronic) power supply of the polarizer magnet. The experiments were, therefore, carried out by moving the depolarizing shim back and forth from a position in front of the counter to a position of small field between the two electromagnets (see Fig. 5), with the two main fields parallel in one experiment and antiparallel (with the rapid reversal) in the next experiment.

The demagnetizations were carried out from 15,400 gauss and 1.25°K, and after lowering the sample tube, a field of ~8000 gauss was slowly applied to the In sample. The counting rate in these experiments was quite high (~32,000 counts/min), but the neutron intensity striking the sample was about 50 times lower than in the case of the open beam experiments because of the attenuation of the Fe block. The counting rate was large,

of course, because no diffraction from an Fe_3O_4 crystal was involved.

Results

A change of 0.50 ± 0.12 percent was observed in the case of parallel fields and 0.4 ± 0.2 percent in the case of antiparallel fields. These changes were in opposite directions, as expected, and were in the direction which would indicate an angular momentum $J = I + \frac{1}{2}$ as before. A total of 10^6 counts of each type (polarized and depolarized neutrons) was obtained. In addition, it was demonstrated that there was no effect within 0.2 percent at 1.2°K under conditions which were otherwise the same as at the low temperatures. It should be mentioned that the smaller effect observed for antiparallel fields should be expected on the basis that the "flipping" efficiency of the abrupt field reversal had been measured to be about 85 percent previously.

A calculation of the effect expected was carried out as follows: If the incident neutron density on the Fe block is given by a Maxwellian distribution with a characteristic temperature of 495°K , the emerging distribution has been given by Stanford⁶⁷ in his Fig. 17 in graphical form. In addition, in his Fig. 18, Stanford⁶⁷ has given the polarization of the emerging neutrons. If we call these $n(v)$ and $f_n(v)$ respectively, and extrapolate the neutron density using the Maxwellian distribution $n(v) = K v^2 e^{-v^2/v_0^2}$ for energies $E \leq 0.17$ eV and $n(v) = C/E$ for energies $E \geq 0.17$ eV, as suggested by Hughes⁷⁰, and extrapolate the neutron polarization by $f_n = F/E$ above velocities of 10^4 cm/sec, which is the upper

limit given by Stanford⁶⁷, we may write in suitable approximation

$$\text{Shim effect} = \frac{C_+ - C_0}{C_0} = \frac{\frac{I}{I+1} \int_0^\infty \alpha(v) n(v) v e^{-Nt\sigma_0(v)} Nt \sigma_0(v) f_n(v) dv}{\int_0^\infty \alpha(v) n(v) v e^{-Nt\sigma_0(v)} dv} \quad (58)$$

where C_+ is the count with polarization, C_0 is the depolarized neutron count, I is the nuclear spin quantum number ($J = I + \frac{1}{2}$ is assumed), f_N is the nuclear polarization, $\alpha(v)$ is the counter sensitivity as a function of velocity, $n(v)$ is the neutron density emerging from the polarizer, v is the neutron velocity, $Nt\sigma_0(v)$ is the macroscopic cross section of the sample as a function of velocity, and $f_n(v)$ is the neutron polarization as a function of velocity.

These integrations were carried out numerically, using the extrapolations and Stanford's data mentioned above, with the result that for a temperature of 0.050°K, the observed effect of 0.5 percent would be obtained. This gave a great deal of confidence that the experiments were progressing satisfactorily, and that it was justified to continue to the case of high neutron polarization.

Experiments IV

These experiments were carried out in February and March 1955.

Neutron Arrangement

After some months of development work, the neutron arrangement

described in Chapter IV and shown in Fig. 6 was obtained. The neutrons diffracted from the Fe_3O_4 crystal were diffracted again from the 111 planes of Cu, thus producing a beam of 87 percent polarized neutrons of energy 0.075 eV which contained only 1.7 percent of second order. These neutrons passed through the indium sample at an angle of approximately $8^{\circ}43'$ from normal incidence. This increased the effective thickness by a small amount, but had no effect otherwise.

Low Temperatures

Demagnetizations were carried out once from 1.25°K and 15,400 gauss, and on four subsequent occasions from 0.99°K and 16,400 gauss. In addition, two duplicate runs at 4.2°K were interspersed with the low temperature runs. The initial temperature less than 1°K was obtained with the aid of the metal booster pump on the liquid helium bath. The procedure in the demagnetizations was fairly standard. The magnet was turned on and stabilized at the desired field value after the He bath had been pumped to a stable pressure (~ 0.11 mm Hg in the last four low temperature runs), the sample tube being in its upper position. After 15 minutes, the valves leading to the 300 liter/sec diffusion pump were opened, and the exchange gas in the sample tube pumped for about 30 minutes. The demagnetization was then carried out by carefully reducing the excitation on the motor generator set supplying the magnet by a factor of about 3, then removing power to the motor so that the set coasted to a standstill. This procedure required some 8 min. The sample tube valves were then closed, and the

sample tube slowly lowered to its lower position; this required about 4 min. The generator was then switched out of the magnet circuit and a bank of eight 12 volt batteries switched in through a rheostat arrangement of a 54 ohm student resistor and three 1-ohm rheostats in series. Enough resistance was removed from the 54 ohm resistor to bring the field to a value just below that required to cause the superconducting transition in indium to occur (270 gauss); then one of the one ohm rheostats was decreased slowly to enable the least possible overshoot in traversing the transition. The remainder of the 54 ohm resistor was then removed, followed by the two remaining one ohm rheostats. The entire bank of resistors was then shorted with a large knife switch. This procedure required about 10 minutes to complete. Zero time for each run was taken as the time when the shorting switch was closed.

Neutron Counting

Counting was started immediately after the full field of 11,150 gauss was applied to the nuclei. Alternating counts of 5 minutes duration were taken, first with the Fe_3O_4 magnet current in the "reverse" direction and with appropriate direct currents through the air core coils to "transmit" the spins of the neutrons unchanged with respect to the field, then with the Fe_3O_4 magnet current in the "forward" direction and the predetermined currents for best "flipping" of the neutron spins passing through the air core coils. These counts may be designated as "R" and "F" counts, these being the initials associated with the direction of the Fe_3O_4 magnet

current. The counting rate was approximately 1000 counts/min, and the background counting rate about 100 counts/min. Background counts were taken before the demagnetization (to save as much counting time as possible at the low temperatures) by keeping the sample tube in its lower position until all preparations (precooling, transfer of liquid He, pumping the He bath to $\sim 1^\circ\text{K}$) for the magnetization of the cooling salts were complete. The background counts were made with both the Fe_3O_4 magnet and low temperature magnets off; this introduced a small error (~ 3 percent) into the background rate because of the slightly different diffracted intensity from the Fe_3O_4 in the two cases. This error is neglected here. With one exception, all F counts made at the low temperature were smaller than the adjacent R counts. The exceptional count was much more than four times outside the average deviation of all other counts and was, therefore, discarded and replaced by the average of several adjacent counts of the same type, which agreed with each other quite well.

The interspersed runs at 4.2°K were exactly the same as those at the low temperatures with respect to the manipulation of the magnetic fields during the neutron counting. In addition to these runs a large number of alternate neutrons counts of the above type were taken at 77°K and at room temperature with the In sample removed from the diffracted beam. The magnetic fields were manipulated in the same way as above in these counts.

Results

At the very low temperatures, all runs made using the same initial conditions gave essentially the same results. If we calculate the percentage difference between the sums of all R and F counts for each run, corrected for background, we obtain the results shown in Table II. It is seen that very reproducible results were obtained in runs 2, 3, 4, and 5. It is also to be noted that the effect in run 1 is lower than in the others because of the higher temperature of the cooling salt expected from the initial conditions.

If we now consider the possibility of observing a time dependence of the effect, and for each of the last four runs, take the sum of all first counts (R type), second counts (F type), third counts (R), etc, and calculate the differences between adjacent counts, corrected for background, we obtain, on averaging adjacent differences, the graph of Fig. 9. The dotted line of this figure is only for the purpose of showing the type of time dependence which might, on general grounds, be expected to occur. An initial rise might be expected as transient heating dies away and a subsequent decrease must occur for warming up of the cooling salt because of steady heating. However, it is emphasized that the data of Fig. 9 are not inconsistent with a very rapid rise (requiring a few seconds) and an exceedingly slow decay, as calculated in Chapter III. The errors shown in Fig. 9 are standard deviations.

The neutron beam from the reactor was not monitored; however, the reactor was controlled during all runs by an automatic regulator based on an ion chamber buried in the reactor only a few feet from the beam hole.

TABLE II

RESULTS OF EXPERIMENTS IV

Run No.	Initial Conditions for Demagnetization	Average Effect, Percent	Overall Average Effect Percent
1	15,400 gauss at 1.24°K	4.95 ± 0.60	5.50 ± 0.30
2	16,400 gauss at 0.99°K	5.44 ± 0.64	
3	16,400 gauss at 0.99°K	5.33 ± 0.72	
4	16,400 gauss at 0.99°K	5.76 ± 0.75	
5	16,400 gauss at 0.99 K	5.45 ± 0.73

Fig. 9. Results of Experiments IV. Average of 4 Runs.

That this control was adequate was substantiated by the runs which demonstrated that the diffracted beam was independent of the neutron spin orientation. These experiments were carried out at Dewar temperatures of 77°K and 300°K, the sample tube being raised to midposition so that neither the sample nor the cooling salt was in the beam. Counts taken in exactly the same way as before showed a difference of only -0.33 ± 0.28 percent, the sign being relative to the low temperature effect.

In order to demonstrate that the effects of Table II were indeed due to the nuclear polarization, two runs (taken after runs 2 and 5 of Table II) were made with the nuclear sample in the beam at 4.2°K. The counting was performed in exactly the same manner as at the low temperatures, with the same magnetic fields. The results of these runs may be summarized by stating that the percentage change observed for all counts at 4.2°K was 0.035 ± 0.52 percent, where a theoretical effect of 0.07 percent was expected. We, therefore, may state that the effects observed in runs 1 - 5 were due to the nuclear polarization.

CHAPTER VII

DISCUSSION OF RESULTS

Direction of Effects

The predominant spin direction for thermal neutrons transmitted by magnetized Fe has been calculated by Schwinger⁷⁶, and measured by Sherwood et al⁷⁷ to be opposite to the direction of the field applied to the Fe. Similarly Sherwood et al⁷⁷ have shown, in a comparison with Fe, that the dominant spin direction in diffraction from magnetized Fe_3O_4 (in first order from the 220 planes) is opposite to the magnetizing field.

Since the magnetic moment of In^{115} is known to be positive³⁶, the energy in a magnetic field corresponding to Eq. (3), $E = -\mu H$, is lowest when μ and H are parallel, and the direction of $\bar{I}_z$ for the polarized In^{115} nuclei is parallel to the field applied to the nuclei.

We thus should expect that the direction of the change in the transmitted intensity, if $J = I + \frac{1}{2}$ is the angular momentum of the compound nucleus in the level corresponding to the most important resonance in In^{115} at thermal neutron energies, would be as in column 5 of Table III, where a comparison is made with the observed direction.

⁷⁶J. S. Schwinger, Phys. Rev. 51, 544 (1937).

⁷⁷J. E. Sherwood, T. E. Stephenson and S. Bernstein, Phys. Rev. 96, 1546 (1954).

TABLE III

DIRECTION OF EFFECTS

(1)	(2)	(3)	(4)	(5)	(6)	(7)
Experi- ments	Polarizer	Analyzer	Neutrons	Expected Direction of Effect (Counting Rates) for $J = I + \frac{1}{2}$	$J = I - \frac{1}{2}$	Observed Direction of Effect
I	In	Fe_3O_4	transmitted	$P > D$	$D > P$	-
II	In	Fe_3O_4	transmitted	$P > D$	$D > P$	$P > D$
IIIa	Fe	In	transmitted	$P > D$	$D > P$	$P > D$
IIIb	Fe	In	flipped	$D > P$	$P > D$	$D > P$
IV R	Fe_3O_4	In	transmitted	$P > D$	$D > P$	$R > F$ (No D counts)
IV F	Fe_3O_4	In	flipped	$D > F$	$F > D$	

P corresponds to counting rate with polarized neutrons.

D corresponds to counting rate with depolarized neutrons.

R, F correspond to "transmitted" and "flipped" neutrons, respectively.

J-value

The J value for the compound nucleus is, therefore, established as

$$J = I + \frac{1}{2}.$$

This result disagrees with that of Brockhouse³³ for the 1.456 eV resonance, which was obtained from determinations of the ratio of scattering to absorption cross sections σ_s/σ_a . In the present experiments it can be shown that the only important resonance is at 1.456 eV. We consider the ratio of absorption cross sections at thermal energies due to the two lowest lying resonances in In¹¹⁵. This may be obtained by assuming that these resonances (hereafter designated as 1 and 2) do not interfere appreciably with one another and may be represented by the Breit-Wigner one-level formula³²

$$\sigma_r = g \pi \lambda \lambda_o \frac{\Gamma_{n_o} \Gamma_r}{(E - E_o)^2 + (\Gamma_n + \Gamma_\gamma)^2/4} \quad (59)$$

where the subscript r refers to the process (resonance scattering or absorption with γ emission), g is the statistical weight factor given by

$$g = \frac{1}{2} \left(1 \pm \frac{1}{2I + 1}\right) \text{ for } J = I \pm \frac{1}{2} \quad (60)$$

for S wave neutron processes, $2\pi\lambda$ is the neutron wave length, E is the energy, Γ_q is the resonance width associated with the emission of the product q of the process, and the subscript o denotes the value at exact resonance. Although we may be dealing with a sum of several like processes, we shall treat the problem as if only one process of each type occurs.

We note also that $\Gamma_n = \Gamma_{n_0} (E/E_0)^{1/2}$. The ratio of absorption cross sections σ_a due to 1 and 2 at thermal energies is then given, in excellent approximation, by

$$\frac{\sigma_a^{(2)}}{\sigma_a^{(1)}} = \left(\frac{E_0^{(1)}}{E_0^{(2)}} \right)^{5/2} \cdot \frac{g_2 \Gamma_{n_0}^{(2)} \Gamma_\gamma^{(2)}}{g_1 \Gamma_n^{(1)} \Gamma_\gamma^{(1)}} = \frac{(\sigma_0 \Gamma_0^2)_2}{(\sigma_0 \Gamma_0^2)_1} \cdot \left(\frac{E_0^{(1)}}{E_0^{(2)}} \right)^{3/2} \quad (61)$$

Landon and Sailor⁷⁸ have recently studied the two lowest energy resonances in elemental indium, using a crystal spectrometer having a resolution of 1.6 min. of arc. Their results are quoted in Table IV. Values of Γ_γ were calculated, assuming $g = \frac{1}{2}$. We have calculated the value in parentheses from other data in the table. The values shown in Table IV were obtained by making a careful analysis of the resonances, including the correction for the Doppler effect^{79,80,81}; an example of the magnitude of this correction is seen from the observed values $\sigma_0^{(1)} = 29,500$ barns and $\Gamma^{(1)} = 109$ mV.

Neutron Width

Using the values of Table IV and Eq. (61), we calculate that

⁷⁸H. H. Landon and V. L. Sailor, BNL 2181 (unpublished).

⁷⁹H. A. Bethe and G. Placzek, Phys. Rev. 51, 462 (1937).

⁸⁰W. E. Lamb, Jr., Phys. Rev. 55, 190 (1939).

⁸¹H. H. Landon, Phys. Rev. 94, 1215 (1954).

TABLE IV

PARAMETERS OF INDIUM 115 RESONANCES

Energy eV	$\sigma_0 \Gamma_0$ barn - eV	σ_0 barn	Γ_0 mV*	Γ_γ mV	$\sigma_0 \Gamma_0^2$
1.456	2880 ± 50	38500 ± 1000	75 ± 2	72 ± 2	216
3.85	103 ± 4	1270 ± 40	81 ± 4	81 ± 4	(8.33)

*mV = 10^{-3} eV in the table headings

$$\frac{\sigma_a^{(2)}}{\sigma_a^{(1)}} \text{ (thermal)} = 0.00606 \quad (62)$$

which demonstrates that measurements of the type carried out in our experiments in the thermal region are equivalent to measurements at the exact resonance energy of 1. The establishment of $J = I + \frac{1}{2}$ also determines a value of g (Eq. (60)) as 11/20 rather than 9/20. Using this value we may calculate $\Gamma_{n_0}^{(1)}$ from the data of Table IV and the equation

$$\Gamma_{n_0} = \frac{\sigma_0 \Gamma_0}{4 \pi g \lambda^2} \quad (63)$$

In this way we obtain

$$\Gamma_{n_0}^{(1)} = 0.00307 \pm 0.00005 \text{ eV} \quad (64)$$

The error being based on the quoted (absolute) error in $\Gamma_0^{(1)}$ of Table IV.

Thermal Absorption Cross Section

Using the center value of Table IV for $\Gamma_0^{(1)}$ (i.e., 0.075 eV), the g value determined from $J = I + \frac{1}{2}$, and the value for $\Gamma_\gamma^{(1)}$ obtained from $\Gamma_0^{(1)} - \Gamma_{n_0}^{(1)}$, the value of the thermal absorption cross section may be calculated from Eq. (59). At 0.075 eV, this value is calculated to be 121.78 barns, and at 0.025 eV, a value of 196.46 barns is obtained. These must be multiplied by the factor 1.00606 from (62) to obtain the values 122.6 ± 6 barns at 0.075 eV for the combined effect of both resonances and

197.6 ± 10 barns at 0.025 eV. This latter value may be compared with the results of Pomerance⁸², who reports a thermal absorption cross section of 191 ± 5 barns based on an assumed value for the thermal (2200 m/sec) absorption cross section for gold of 95 barns. A revision to 98.7 ± 0.6 barns for this value⁸³ raises the Γ_n value from Pomerance's data to 198.4 ± 6 barns. The agreement is excellent.

Width Ratio

If we calculate Γ_γ by subtracting the value of Γ_{n_0} found from (63) and given in (64) from the center value for Γ_0 , and then take the ratio $\Gamma_{n_0}/\Gamma_\gamma$, the result is

$$\Gamma_{n_0}/\Gamma_\gamma = 0.041 \pm 0.0013 \quad (65)$$

This is to be compared with the result of Brockhouse³³ who obtained a value 0.043 ± 0.006 from determinations of σ_s/σ_a as a function of energy. The agreement is excellent. It is unfortunate that the data which he used⁸⁴ for the resonance parameters, namely $\sigma_0 = 27,000$ barns, and $\Gamma_0 = 110$ mV, had not been corrected for the Doppler effect, or were not corrected to a sufficient extent, and this led to his incorrect conclusion that $J = I - \frac{1}{2}$ for the first resonance.

⁸²H. Pomerance, Phys. Rev. 83, 641 (1951).

⁸³Carter, Palevsky, Myers and Hughes, Phys. Rev. 92, 716 (1953).

⁸⁴V. L. Sailor and L. B. Borst, Phys. Rev. 87, 161 (1952).

Magnitude of Effect in Experiments IV

The size of the effects observed in Experiments II and III have already been discussed in Chapter VI to the extent to which such discussions are merited. It is felt that a more complete discussion of the effects observed in Experiments IV is in order here.

While the general agreement of the calculation of the temperature of the indium with the temperature derived from the nuclear effect observed is fairly satisfactory, certain discrepancies should be discussed. If the calculations of the time constants (for both cooling and warm-up) which were carried out in Chapter III are substantially correct, the temperature variation during an experiment would be rather small, and the time independent average of the effect observed should be used to calculate the temperature of the indium. This was $0.054 \pm 0.003^{\circ}\text{K}$ based on the average effect obtained in the last four runs, and is substantially higher than either of the calculated temperatures (47) or (49). As may be seen from the form of Eq. (45), the assumed thermal conductivity of the silver wires or indium plates would have to be decreased considerably to appreciably raise the calculated temperatures. Alternatively, using the Oxford data, the assumed heat input rate would have to be increased by a substantial factor (~ 3) to account for the "observed" temperature mentioned above. Under this latter condition the temperature increase during an experiment would be expected to be still rather small, namely, about 0.002°K , which would produce a decrease in the neutron effect of only

5 percent during this time. Using the Leiden data, an assumed heat input of 120 erg/min would bring about an initial temperature of $\sim 0.038^{\circ}\text{K}$ and a final temperature of $\sim 0.058^{\circ}\text{K}$. This would be in fairly good agreement with the temperatures calculated from that portion of the dotted curve of Fig. 9 after $t = 30$ min. This is not to say that any particular validity is attached to the dotted curve of Fig. 9, but that the curve represents a possible behavior, and subjectively appears to be a good representation.

The initial rise of this curve could possibly be explained on the basis of the presence of the guard cage. This cage was made of a Cu tube which had been partially milled away, leaving strips of metal ~ 3 mm wide by 0.5 mm thick in a rectangular network on the cylindrical surface with spacings of about 1.5 cm around the periphery and 5 cm along the axial direction. Substantial eddy current heating occurred in this cage. A rough evaluation of the time constant for cooling, in the manner of the analysis of Eq. (52) gave extremely long thermal relaxation times at temperatures $< 0.1^{\circ}\text{K}$ for the salt, and times of ~ 10 min. at temperatures of $\sim 0.2^{\circ}\text{K}$. In addition, a 10.6 minute time constant for cooling was once observed with this sample assembly (from salt susceptibility measurements) for both coolant salts after suddenly switching a magnetic field off while the nuclear sample was in the magnet gap. It is postulated that a rise such as that in the dotted curve of Fig. 9 could be caused by the upper end of the cage being heated substantially by eddy current heating during the nuclear magnetization, then cooling very slowly because of the rel-

atively poor heat contact to the cage salt. The surface contact area was $\sim 9 \text{ cm}^2$, but the distance from the metal cage to the center of the salt pill was $\sim 1 \text{ cm}$, in contrast to the much shorter maximum distances from metal to salt ($\sim 0.05 \text{ cm}$) prevalent in the salt cooling the indium. The cage contained the equivalent of $\sim 0.5 \text{ mol}$ of Cu, and a rough calculation indicated that the eddy current heating received in the last step of the nuclear magnetization might easily have been sufficient to raise the metal temperature above 1°K . It should be mentioned that this is in agreement with the fact that the cage salt temperature (as determined from susceptibility measurements) was always a good bit higher than that of the inner salt throughout the runs. Thus as quasi-equilibrium conditions were approached (at around 0.2°K) between the cage and its salt, the possibility exists that there was at the upper end of the cage, a variable temperature source of heat to the indium plates through their mounting. This would appear to be the only plausible explanation for the 10 minute relaxation time observed at these temperatures for both salts, and the possibility of a rise time for the nuclear effect of about this magnitude.

It is also possible, since the temperature of the cage salt rose substantially during the course of a run, that a decrease in the effect after $t = 30 \text{ min.}$ could be accounted for by the warm-up of the cage assembly which would tend to increase the heat current through the plastic supports, and cause a larger temperature drop between the indium plates and the inner salt, as well as a rise in the inner salt temperature itself. This is not considered to be quite as likely an explanation as that above for

the initial rise in the effect.

It is also clear from the discussions given above that no conclusions may be drawn regarding the correctness of either the Oxford or Leiden data on the low temperature properties of $\text{Fe}(\text{NH}_4)(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$, and that considerable work would be required to completely determine the situation with regard to heat transfer.

Possibilities for Future Development

The calculations and estimates which have been given indicate three distinct improvements which may be made in future experiments directed toward obtaining larger polarizations, perhaps with the intention of performing a nuclear demagnetization. First, it is certain that no fundamental difficulty would appear in increasing the magnetic field applied to the nuclei, certainly by a factor of ~ 3 . Secondly, in view of previous discussions, it is somewhat surprising to find that the limitation to the temperature attainable was apparently the metallic conduction problem rather than conduction within the salt. This immediately indicates that extremely pure, annealed, and carefully handled (to prevent cold-working) metallic components should be used in future experiments. Thus, substantial reductions in the temperature might be obtained, although temperatures $< 0.01^\circ\text{K}$ may not be practically possible. It should be noted that the electrical resistivities of such metal samples would be smaller, and the problem of eliminating eddy current heating more difficult. Third,

arrangements for preventing heat leaks, and transient heating may be improved substantially over the arrangement here, especially with regard to eddy current heating in the cage.

One may conclude that a nuclear demagnetization appears to be a possible experiment, provided that the means of isolating the nuclear sample thermally after polarizing the nuclei can be developed, and that the heat leaks can be made sufficiently small.

Our suggestion of the "foil" method of flipping polarized neutron spins with respect to the magnetic field may be of use in a) flipping neutrons of very low energy because of the strong gradients of field made possible, and b) in high energy experiments where polarized neutrons may be produced using nuclear reactions. The method may offer an advantage in overcoming, with the aid of deliberately introduced steady field components, undesired effects of small stray field gradients.

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